

NONLINEAR OPTICS OF NEMATIC LIQUID CRYSTALS

Dissertation
submitted to the
Fakultät für Physik
Ludwig-Maximilian Universität
München

by

Fabienne Marquis
from Sion (Switzerland)

München, January 8th. 1987

1. Gutachter : Prof. P.Meystre

2. Gutachter : Prof. H.Wolter

Tag der mündlichen Prüfung : 26. Februar 1987

ABSTRACT

In this thesis, we study the molecular reorientation induced by a laser beam in a thin sample of nematic liquid crystal. For laser intensities larger than the threshold of the optical Freedericksz transition, molecular reorientation occurs and induces a nonlinear variation of the refractive index of the medium, which in turn modifies the light propagation through the liquid crystal. In the case of small dielectric anisotropy, a one-dimensional model of the molecular reorientation can be solved analytically and leads to a mode structure of the angle distribution characterizing the reorientation. A dynamical analysis shows that only the lowest-order mode of the reorientation is stable. When transverse elastic correlations are considered between the molecules, a two-dimensional model is developed for the small reorientation regime. It is shown for this case that inhomogeneous transverse profiles of the reorientation are possible, even under plane wave pumping. In the general case of arbitrary laser intensity and dielectric anisotropy, a numerical solution of the full two-dimensional problem for the case of a gaussian transverse profile of the laser intensity confirms the mode structure of the molecular reorientation on the longitudinal dimension and leads to a similar mode structure for its transverse dependence. The fundamental mode is the only nonhomogeneous solution for the angle distribution for a laser intensity close to threshold. When the intensity is further increased, each higher mode becomes successively possible. A numerical study of the dynamical system shows that the fundamental mode is the only stable one. Critical slowing down is exhibited close to threshold, characteristic of the second-order phase transition occurring at the Freedericksz threshold. A different approach considers the thermal fluctuations present in the medium and develop a stochastic description of the molecular reorientation. Performing a numerical simulation of the noise shows that transitions to higher modes of the reorientation can be induced by thermal noise. Finally, we consider an application of our model by discussing the case of a Fabry-Perot resonator containing a nematic liquid crystal. Bistable output of the resonator is exhibited, due to the light-induced molecular reorientation in the nonlinear medium. This optically bistable system is somehow different from more conventional systems, due to the form of the nonlinearity that leads to a quadratic dependence of the transmitted intensity on the input intensity.

OVERVIEW

I. INTRODUCTION

- I.1 Liquid crystals and Freedericksz transition
- I.2 Previous results
- I.3 Overview

II. CONTINUOUS THEORY OF THE NEMATIC LIQUID CRYSTAL

- II.1 Free energy
- II.2 Minimization of the free energy - homeotropic boundary conditions
- II.3 Discussion

III. TWO ANALYTICAL APPROXIMATIONS : SMALL DIELECTRIC ANISOTROPY AND SMALL REORIENTATION REGIME

- III.1 Small dielectric anisotropy
- III.2 Two dimensional model, small reorientation regime
- III.3 Effect of the finite spot size of the laser beam

IV. NUMERICAL SOLUTION OF THE DIRECTORS EQUATION

- IV.1 Steady state
- IV.2 Dynamics

V. STOCHASTIC DESCRIPTION OF THE OPTICAL FREEDERICKSZ TRANSITION

- V.1 Introduction
- V.2 Langevin approach - Numerical solution
- V.3 Fokker-Planck approach

I. INTRODUCTION

VI. OPTICAL BISTABILITY OF A FABRY-PEROT RESONATOR CONTAINING A NEMATIC LIQUID CRYSTAL

VI.1 Optical bistability

VI.2 Bistability near the optical Freedericksz transition

VII. SUMMARY AND OUTLOOK



Fig. 1.1. Molecular orientation in a nematic liquid crystal, the director is defined as the unit vector of the average.

I. INTRODUCTION

I.1 LIQUID CRYSTALS AND FREEDERICKSZ TRANSITION

Some organic compounds exhibit between their solid and isotropic liquid phases a mesophase described at the end of last century by Lehmann [Lehmann 1890] as "liquid crystal". This liquid crystalline phase corresponds to a state where matter is fluid, but where the molecules are arranged with a certain order. The isotropy of a liquid is broken, and the spatial regular order of a crystal is not present, so that the state is "intermediate" between liquid and crystal. Several mesophases have been observed, involving different types of order. In a smectic phase, the molecules exhibit a one dimensional order, forming layers separated by a well defined mean distance. In each layer, however, the medium has no spatial order: rather, it has the characteristic of a true liquid. In the nematic mesophase, the liquid is composed of rod-like or disk-like molecules, which are on the average aligned parallel to a certain preferential direction, but with no positional order of their centers of gravity.

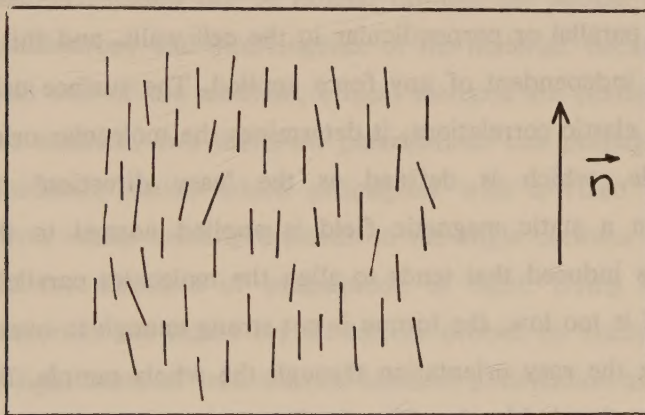


Fig. 1.1 Molecular orientation in a nematic liquid crystal, the director $\mathbf{n}$ defines the mean orientation of the medium.

This class of liquid crystals can be seen as a particular case of cholesteric liquid crystals: in this third class, parallel layers of nematic compounds having different orientations are stacked on each other, so that the mean orientation follows a helicoidal structure perpendicularly to the layers, with a well defined helix pitch. A cholesteric of infinite pitch is just a nematic.

The electric and magnetic anisotropy of liquid crystals and the tendency of the molecules to follow the lines of force when placed in a magnetic field were discussed as early as 1900 [Lehmann 1900]. Since then, numerous works have provided a better understanding of this intermediate state of matter, and in particular of its interaction with electric and magnetic fields, its thermic and elastic properties.

In 1927, Freedericksz and Repiewa [Freedericksz and Repiewa 1927] observed that a liquid crystal placed in a magnetic field experiences a collective reorientation of its molecules if the magnetic field is higher than a threshold value. This effect is known as the Freedericksz transition.

A nematic liquid crystal (NLC) placed in a cell is characterized by a macroscopic orientation of its molecules, defined by the orientation at the surfaces of the cell. Different factors determine the surface orientation, such for example the chemical structures of the liquid crystal and the cell material, or the way the cell surfaces are treated. To observe the Freedericksz transition, the orientation at the surface has to be parallel or perpendicular to the cell walls, and this anchoring must be strong, ideally independent of any force applied. The surface orientation is then fixed, and due to elastic correlations, it determines the molecular orientation through the whole sample, which is defined as the "easy direction" of the medium (Fig. 1.2(a)). When a static magnetic field is applied normal to this easy axis, a magnetic torque is induced that tends to align the molecules parallel to the field. If the magnetic field is too low, the torque is not strong enough to overcome the elastic forces maintaining the easy orientation through the whole sample. But for magnetic fields larger than a threshold value H_C , the Freedericksz transition occurs and aligns the molecules along H in the bulk of the crystal. At the surface, the strong anchoring maintains the initial orientation along the easy direction and a transition region leads continuously from one orientation to the other. For values of the field just above H_C , this transition region is broad and the distortion from the easy axis is weak, as illustrated in Fig. 1.2(b). For fields much higher than H_C , the transition is

very sharp and a total alignment with the field is achieved through the whole sample except in thin transition regions near the surfaces (Fig. 1.2(c)). Note that the Freedericksz transition can be induced by an electric field as well as by a magnetic field.

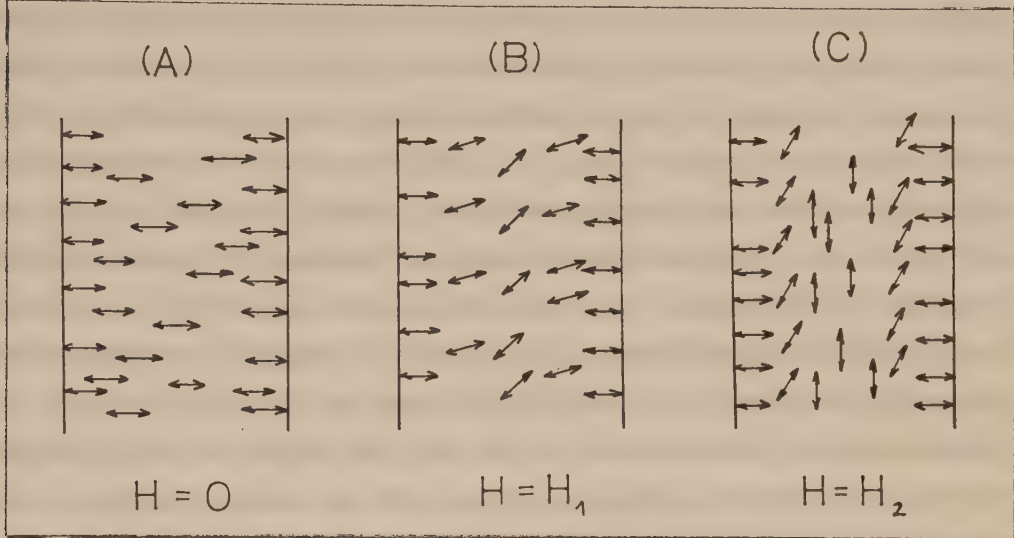


Fig 1.2 Molecular reorientation induced by a static magnetic field in a NLC. (a) H is under threshold; (b) H_1 is just above threshold; (c) $H_2 \gg H_c$

The molecular orientation can be probed through the sample in different ways, using the strong anisotropy and birefringence of the material. Because the orientation defines the optical axis of the medium, optical methods are particularly well suited. In a birefringent medium, two different polarizations can propagate without being perturbed: the ordinary wave, which propagates with a fixed velocity, and the extraordinary wave, whose velocity depends on the angle between the optical axis of the medium and the direction of propagation of light. Using this property, the molecular orientation in the liquid crystal can be probed by studying the change in polarization of a light beam of well defined incident polarization as it travels through the medium. For example, a NLC perpendicularly aligned along the z -axis is seen as an isotropic structure by input light linearly polarized in the x - y plane and perpendicularly incident on the medium. When reorientation occurs in the x - z plane, the x component of the polarization propagates with an orientation-dependent velocity, while the y component of the polarization has constant velocity. The output light is probed through an analyser oriented at $\pi/2$ with respect to the input

polarization. When the molecules are uniformly aligned on the z direction, no light is transmitted through the analyser, but transmission occurs when the molecules are reoriented, [Goodman 1974], and is a function of the reorientation. Conoscopic methods [De Gennes 1975] to probe the orientation of a sample use converging polarized light illuminating the medium, and study the distortion of the interference pattern. When the medium is unperturbed with all the molecules aligned along the preferential direction, a linearly polarized beam converging on the liquid crystal forms at the output plane a symmetric interference pattern of concentric dark and bright circles. When the molecular orientation is rotated, the circles become distorted and the distortion is a function of the angle of reorientation. Another possible way to measure the reorientation is to add some impurities in the NLC [Goodman 1974], such as dichroic dye molecules, characterized by an absorption spectrum which is a function of the orientation of the molecules with respect to the polarization of the incident light. In a nematic host, the dye molecules follow the main orientation of the medium, and their optical axis rotate with the nematic directors when the Freedericksz transition occurs. Different absorption coefficients then characterize the NLC containing dye impurities when the probe light is polarized parallel or perpendicular to the main optical axis. When a polarized white light probes the sample of NLC, the color of the outcoming light can be used to determine the orientation in the sample. [Kahn 1972]

To treat collective phenomena occurring on a scale larger than the molecular dimension, a continuum theory of NLC was developed [Zocher 1933; Oseen 1933; Frank 1958]. Using a macroscopic theory, a general form of the elastic energy of the liquid crystal can be written, and the orientation of the molecules submitted to external forces can be determined as the one minimizing the free energy of the medium.

In the Freedericksz transition induced by a static magnetic field, the field induced reorientation modifies the propagation of probe light beams traveling through the medium. The same process can be totally optical, an electromagnetic field being then responsible for the same effects as the static field. In the optical Freedericksz transition (OFT), the reorientation is directly induced by an optical field incident on the NLC, and the same light responds to the change of the optical properties induced in the medium by molecular reorientation. An important difference must be considered between the Freedericksz transition induced by a

static magnetic field, where the field is constant, and the OFT, for which on the contrary a polarization field is induced in the sample, implying that the electromagnetic field in the medium is function of the reorientation.

In the present thesis, we use the continuum theory to study the optical Freedericksz transition, consisting of the molecular reorientation of a NLC due to the effect of a perpendicularly incident optical field. This transition has been intensively studied since the beginning of this decade, both theoretically and experimentally. Our model consists of a sample of NLC in the homeotropic alignment, i.e. characterized by an easy direction perpendicular to the cell walls. Its longitudinal dimension is d and its transverse dimension is taken to be infinite. The illumination is perpendicular to the cell walls, and is produced by a linearly polarized laser beam. The main direction of orientation of the molecules, described by a vector $\mathbf{n}$, is homogenous and perpendicular to the surfaces in the state unperturbed by the laser light. When the laser is switched on, electromagnetic forces compete with elastic forces due to the strong anchoring at the surfaces, and the molecules rotate to a position that minimizes the free energy of the sample.

This work is motivated by the fact that the OFT occurring in a nematic medium is a good candidate to study light induced nonlinear processes. The giant optical nonlinearity of the nematic phase allows to study nonlinear light induced effects using CW laser beams of reasonable intensity. A theoretical model of the OFT can be developed in a relatively simple frame, which under some approximations is amenable to analytical treatment. From the experimental point of view, a number of different nematic compounds have been studied, their characteristic parameters are well known. Since they are liquid media, they are easy to handle experimentally, and the theoretical predictions of our model can be confronted with experimental results.

As will be discussed in this thesis, transverse effects can be included in the description of the OFT and lead to interesting solutions even when treated in simple models. In many problems studied in nonlinear optics, transverse effects play an important role, but often lead to barely manageable complications of theoretical models. The study of the OFT is an example where transverse effects can be considered in a relatively simple way. Finally, liquid crystals are commonly used in practical devices; a broad choice of materials is available, but the richness of their nonlinear optical effects is still not totally explored and can probably lead to interesting novel applications.

1.2 PREVIOUS RESULTS

Various aspects of the OFT have already been studied, under different geometries, both theoretically and experimentally. The theoretical models of the OFT discussed in the literature are based on the continuous theory and describe the light induced molecular reorientation as an effect of competing elastic and electromagnetic forces. The elastic energy is expressed following Franck's description of the elastic distortions in the medium. In the first theoretical analysis of the OFT, Zel'dovich, Tabiryan and Chilingaryan [Zel'dovich et al. 1981; Zel'dovich and Tabiryan 1982] first determine the electric field in the medium by solving self-consistently Maxwell's equations in a medium characterized by a nonhomogenous anisotropic dielectric tensor, and deduce then the electric energy due to the field. They do not consider in their model the contribution of the magnetic part to the total free energy of the liquid crystal. The Euler-Lagrange equation for the directors is obtained by minimizing the free energy for fixed amplitudes of the electric field.

Durbin, Arakelian and Shen [Durbin et al. 1981a] discuss their first experimental observations of the OFT using a theoretical model based on an infinite plane wave approximation involving only a dependence of the reorientation on the longitudinal dimension. Their theory assumes a very broad beam and discusses in a one dimensional problem the reorientation as function of the intensity of the input laser, expressed through a Poynting vector of constant magnitude.

Both approaches to describe the electromagnetic energy are discussed critically by Ong [Ong 1983], who points out that the magnetic energy has to be considered, since it contributes to the total free energy by the same amount as the electric term. Furthermore, the amplitude of the electric field is not constant, but depends on the local director. In his model, Ong assumes a plane wave input beam and suggests that only the magnitude of the z component of the Poynting vector, S_z , is constant since the dielectric tensor varies with position. Expressions for the electric and magnetic fields are then obtained solving Maxwell's equations for a plane wave input beam, and the electromagnetic energy is expressed as a function of S_z , which is a constant under the assumptions that diffraction and absorption are negligible. This model only considers a longitudinal dependence of the reorientation and leads to an implicit analytical form of the reorientation angle, given in the form of an integral equation.

The reorientation angle is discussed in the limit of very broad beams, assuming a solution of the form $\theta(z) = \theta_m \sin(\pi z/d)$, where θ_m is the maximum angle of reorientation and d is the length of the liquid crystal, by Zel'dovich et al. 1981, Ong 1983. This approximation is well suited for small intensities and leads to a threshold intensity proportional to κ_{33}/d^2 , where κ_{33} is the bend elastic constant.

For narrow beams, the transverse dependence of the reorientation angle has to be considered, a one dimensional model is then not well suited. The results of a three-dimensional linearized calculation lead to a transverse profile of the reorientation expressed by a Bessel function of argument $\pi r/d$, r being the transverse coordinate [Csillag et al. 1982; Zel'dovich et al. 1981].

These basic models are helpful in discussing the results observed in several experimental studies of light induced nonlinear effects in NLC. Optical diffraction is observed [Durbin et al. 1982; Khoo 1982; Khoo 1983] when two incoming beams are combined in a thin sample of NLC. Their interferences induce a spatially modulated index of refraction, due to molecular reorientation, responsible for diffraction of the incoming light.

When the Freedericksz transition is induced in a NLC, self phase modulation due to light induced birefringence results in a ring pattern formed by the outgoing light. As many as 100 rings have been observed, [Durbin et al. 1981b], suggesting very large induced nonlinear phase shifts. The induced phase shift has been measured in the first experimental observation of the OFT [Durbin et al. 1981a], and values of the induced nonlinear phase shift as high as $40 \times 2\pi$ have been recorded.

When a nematic liquid crystal is placed in a Fabry-Perot cavity, multistable output can be observed, due to the nonlinearity of the nematic sample and to the feedback provided by the resonator. Numerical experiments confirm this behaviour [Arakelian et al. 1983].

Beside the light-induced molecular reorientation, thermal effects in the medium modify the refractive index and induce a nonlinear phase shift [Shen 1984; Khoo and Hou 1985]. When absorption is present in the medium, the temperature of the sample is increased by laser heating. The index of refraction, which is a function of temperature, is then changed and a thermally induced phase shift is generated in the medium. Laser heating and molecular reorientation are characterized by different

time constants and can lead to competing effects, as was measured in the transient behaviour of a NLC illuminated by a pulsed laser [Hsiung et al. 1984]. The time constants of molecular reorientation and thermal effects were measured and shown to be vastly different, the thermal effects being about 3 orders of magnitude faster than the reorientational effects. When the liquid crystal is placed in a resonator, these two counteracting effects can lead to self-oscillation of the output light intensity [Cheung et al. 1983].

The time constant of light induced molecular reorientation is very large and is a function of the intensity of the incoming light [Durbin et al. 1981a]. It can be of the order of several hundreds of seconds close to threshold, and decreases with increasing input intensities like $(I - I_{th})^{-1}$, where I_{th} is the threshold intensity of the OFT. This behaviour correspond to critical slowing down and is characteristic of a second order phase transition.

The Freedericksz transition is usually of second-order, but for specific values of the medium parameters (elastic constants, dielectric anisotropy), it can become first order and exhibit hysteresis [Ong 1983]. This possibility is discussed theoretically by Ong and has been recently confirmed experimentally [Ong 1985; Karn et al. 1986]. This implies that a NLC illuminated by a laser beam can be an intrinsic bistable system involving no external feedback mechanism.

1.3 OVERVIEW

In this work, a theoretical model of the OFT in a NLC is developed, following Ong's approach to describe the electromagnetic energy. His model is extended to take into account transverse dependence of the molecular reorientation, introducing transverse correlations between the molecules due to the elastic forces acting in the medium. This model is solved to study the longitudinal and transverse distributions of the molecular reorientation. The continuous theory of a nematic is presented in Chapter II : the free-energy $\mathcal{F}$ of the liquid crystal in an electromagnetic field is calculated for a general case. Introducing a specific geometry, $\mathcal{F}$ is minimized under the assumptions that the laser light is neither diffracted nor absorbed in the medium and that the elastic properties of the liquid crystal are isotropic. The dissipative effects due to the viscosity of the medium governing the dynamics of the molecular

reorientation are introduced to derive the equation of motion for the directors.

This theory is used in Chapter III to study two particular cases. In a medium characterized by a small dielectric anisotropy, the threshold intensity of the OFT is calculated in a one-dimensional model, assuming a plane wave incident field, and the longitudinal distribution of the reorientation angle $\theta(z)$ is shown to be described above threshold by an elliptic sine function. As for the trigonometric sine, the elliptic sine sn is a periodic function which exhibits an infinite set of zeros. Different modes of the molecular reorientation can then be defined, characterized by an increasing number of nodes between two fixed zeros of the function. All these modes of the sn can be steady-state solutions of $\theta(z)$ for high enough intensities. The stability of the modes is numerically solved and the fundamental mode is the only stable one. A numerical integration of the dynamical equation is performed, showing that all initial configurations evolve towards the first mode distribution within a time scale inversely proportional to the excess threshold intensity. A second particular case studies the OFT close to threshold, where the molecular reorientation is characterized by small values of θ . A two-dimensional model is considered, using a third-order approximation in θ of the nonlinearity. The solution shows that transverse inhomogeneities of the molecular reorientation are possible even under plane-wave illumination. An analytical study of the dynamics of the transverse reorientation leads to traveling-wave types of solutions. Based on this small θ expansion, the last Section discusses the effect of the finite spot-size of the laser beam, assuming that the laser has a transverse intensity profile described by a quadratic function. The threshold value of the intensity is discussed as a function of the beam diameter.

Chapter IV is devoted to a numerical study of the Freedericksz transition for the case where two spatial dimensions are involved, in the general situation where the full nonlinearity of the medium is considered. The problem is treated to all orders both in the dielectric anisotropy and in the reorientation angle. The transverse profile of the laser intensity is assumed to be gaussian. The numerical solution of the steady-state equation describing the molecular reorientation exhibits a fundamental solution characterized by a maximum reorientation in the center of the sample. However, higher modes of the reorientation are still possible both along the transverse and the longitudinal dimensions. Some of these higher modes are calculated as steady-state solutions of the full numerical problem. The second Section studies the dynamics of these modes; a numerical integration of the time-dependent equations shows that the stable configuration of the molecular reorientation is

described by the fundamental mode. Higher modes are however present in transient regime, and can live for very long times if the intensity of the input laser is large compared to their respective thresholds.

Chapter V considers a statistical description of the molecular reorientation. A stochastic force describing the thermal fluctuations present in a real experiment is introduced, which allows the decay of dynamically unstable solutions. The Langevin equation is solved numerically. We find that due to the presence of thermal noise in the system, higher order modes of the molecular reorientation can now be reached. The equivalent Fokker-Planck equation corresponding to our Langevin description is derived in Section 3, and is used to determine the probability to observe a Freedericksz transition to different modes of the molecular reorientation.

Chapter VI studies the behaviour of a Fabry-Perot cavity containing a nematic liquid crystal. In the first Section, we briefly review the general problem of a resonator cavity filled with a nonlinear medium and show the possibility of bistable regime. This general theory is applied in Section 2 to the case where the nonlinear medium is a NLC and the source of the nonlinearity is light induced molecular reorientation. This system is somehow different from more conventional optically bistable systems, since the nonlinear relation between input and output intensities of the Fabry-Perot is of a quadratic form rather than cubic.

The last Chapter summarizes the main results of this work and concludes with some suggestions for possible future directions in the study of the OFT.

II. CONTINUOUS THEORY OF THE NEMATIC LIQUID CRYSTAL

II.1 FREE ENERGY

The OFT involves a collective behaviour of the molecules: the molecular reorientation induced by the field acts on a space scale much larger than the molecular dimension. For the study of electromagnetic field induced effects, the NLC is described as a continuous medium with specific elastic properties. Such a phenomenological continuous theory of liquid crystals has been developed by Zocher, Oseen and Frank [Zocher 1933; Oseen 1933; Franck 1958].

The medium is described by introducing small macroscopic domains, each of which containing a large number of molecules. In each domain, a main direction of orientation can be determined by averaging the molecular orientations. It is described by a unique director vector $\mathbf{n}$, pointing along this direction. The field of director vectors at each point (a point corresponding to one small domain) defines then the macroscopic orientation of the sample.

The NLC is an elastic medium which can be deformed, inducing a distortion of the director field. In such a distortion, the free energy $\mathcal{F}$ of the medium is modified by the elastic deformation. The explicit form of $\mathcal{F}$ involves the various moduli of elasticity (Hooke's law). Considering the useful symmetry properties of a nematic medium :

- it is a uniaxial medium with cylindrical symmetry about the axis defined by the director vectors,
- in a nonpolar nematic (the medium has no permanent dipole moments), $\mathbf{n}$ and $-\mathbf{n}$ are not distinguishable,
- the medium has mirror symmetry,

the relevant elastic moduli are reduced to three, which are known as the Frank elastic constants [De Gennes 1975]. κ_{11} is the elastic modulus characterizing a splay distortion of the director field, for which $\nabla \cdot \mathbf{n} \neq 0$. κ_{22} and κ_{33} correspond respectively to a twist ($\mathbf{n} \cdot \nabla \times \mathbf{n} \neq 0$) and bend ($\mathbf{n} \times \nabla \times \mathbf{n} \neq 0$) deformation. The elastic constants decrease with increasing temperature. Any possible distortion of the director field of a NLC can be decomposed on these three contributions. The bulk

elastic free energy density of deformation can be expressed as :

$$F_{el} = \frac{1}{2} [\kappa_{11}(\nabla \cdot \mathbf{n})^2 + \kappa_{22}(\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + \kappa_{33}(\mathbf{n} \times \nabla \times \mathbf{n})^2]. \quad (2.1)$$

On the surfaces of the sample, the directors tend to follow an easy direction, depending upon the way the surfaces are treated. There are different ways to prepare the liquid crystal sample so that a preferential orientation is fixed at the walls. For example, a homeotropic alignment (directors aligned perpendicularly to the surface) can be obtained by treating the surfaces of the cell containing the liquid crystal with lecithin or other lipids; these compounds will be perpendicularly fixed at the surface, imposing a homeotropic alignment of the nematic in the cell. It is also possible to align the directors along one preferential direction by simply rubbing the glass surface of the cell in this direction. It is assumed here that the wall-molecules interaction is at least of the same order of magnitude as the molecular-molecular interaction in the bulk, so that no reorientation of the directors at the surface is induced by the molecular reorientation in the bulk. In this case, the surface energy can be neglected, and the only effect of the surface is to impose fixed boundary conditions.

When the NLC is submitted to an external field, a new contribution must be added to the free energy. In the case of an electromagnetic field, it is given by the electromagnetic energy density

$$F_{em} = \frac{1}{2} [\mathbf{E} \cdot \mathbf{D} + \mathbf{B} \cdot \mathbf{H}]. \quad (2.2)$$

In practice, the medium can be treated as nonmagnetic ($\mu = 1$), since the major electromagnetic effect is due to the dielectric anisotropy, which is typically 10^6 times larger than the magnetic anisotropy [Ong 1983]. The dielectric response of the medium $\bar{\epsilon}$ is a function of the angle α between the direction of propagation of light and the optical axis determined by $\mathbf{n}$. Light interacting with the medium produces an electromagnetic torque, which tends to reorient the directors, acting against the elastic torque which tends to uniformly align them along the easy direction determined by the boundary conditions. This induces a change in α , and hence a variation of $\bar{\epsilon}$ which in turn modifies the light propagation in the liquid crystal. This

nonlinear interaction expressed by the electric displacement

$$\mathbf{D} = \bar{\epsilon}(\alpha) \mathbf{E} \quad (2.3)$$

couples the elastic and electromagnetic effects.

These two competing contributions give the total free energy of the liquid crystal

$$\begin{aligned} \mathcal{F} &= \int_V d^3r [\mathcal{F}_{el} - \mathcal{F}_{em}] \\ &= \frac{1}{2} \int_V d^3r [\kappa_{11}(\nabla \cdot \mathbf{n})^2 + \kappa_{22}(\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + \kappa_{33}(\mathbf{n} \times \nabla \times \mathbf{n})^2 - 2 \mathcal{F}_{em}]. \end{aligned} \quad (2.4)$$

The steady-state distribution of $\mathbf{n}$ through the sample for a given input light intensity corresponds to the vector configuration $\mathbf{n}$ that minimizes this free energy.

II.2 MINIMIZATION OF THE FREE ENERGY - HOMEOTROPIC BOUNDARY CONDITIONS

In this Section, we derive the distribution of directors $\mathbf{n}$ which minimizes the free energy, for the case of a homeotropic sample sandwiched between two plates of infinite transverse dimensions at $z = 0$ and $z = d$. In the absence of laser light, the directors are aligned along $\mathbf{e}_z$, the unit vector in the z -direction. We assume the laser to be perpendicularly incident on the crystal surface, with its propagation vector $\mathbf{k}$ parallel to the easy direction $\mathbf{e}_z$, and linearly polarized along the x -direction, as shown in Fig. 2.1.

For this choice of polarization, it is a good approximation to assume that the distribution of directors is stable against fluctuations along the y -direction, so that the directors always point in the x - z plane. But due to the polarized laser field, a reorientation of the directors is possible in this plane. We describe this reorientation

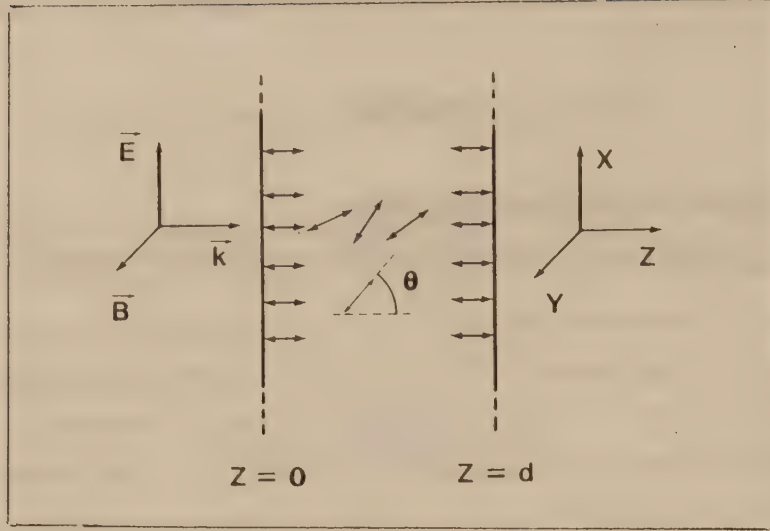


Fig 2.1 Sample of NLC, homeotropically aligned, pumped perpendicularly by a linearly polarized laser beam. θ is the angle between the directors and the z-axis.

by the angle $\theta(x,z)$ between $\mathbf{n}$ and $\mathbf{e}_z$. F_{el} can then be expressed explicitly as a function of θ . With $\mathbf{n} = (n_x, n_y, n_z) = (\sin\theta, 0, \cos\theta)$, we obtain

$$(\nabla \cdot \mathbf{n})^2 = \cos^2\theta (\partial_x \theta)^2 + \sin^2\theta (\partial_z \theta)^2 - 2 \sin\theta \cos\theta \partial_x \theta \partial_z \theta$$

$$(\mathbf{n} \cdot \nabla \times \mathbf{n})^2 = 0$$

$$(\mathbf{n} \times \nabla \times \mathbf{n})^2 = \cos^2\theta (\partial_z \theta)^2 + \sin^2\theta (\partial_x \theta)^2 + 2 \sin\theta \cos\theta \partial_x \theta \partial_z \theta. \quad (2.5)$$

The electromagnetic energy is calculated using Maxwell's equations. We show in Appendix A that for a wave of frequency ω and wave vector $\mathbf{k} = \frac{\omega}{c} \mathbf{n}_p$, where $\frac{c}{n_p}$ is the phase velocity of light in the medium, F_{em} can be expressed as

$$F_{em} = \frac{1}{c} \mathbf{S} \cdot \mathbf{n}_p = \frac{1}{c} S_z n_p, \quad (2.6)$$

where $\mathbf{S}$ is the Poynting vector. Under the approximation that diffraction and absorption of the beam can be neglected, its z-component S_z remains constant

during propagation through the medium [Ong 1983]. The expression of n_p for a birefringent medium whose ordinary and extraordinary indices of refraction are n_o and n_e is [Born and Wolf 1970]

$$n_p = \frac{n_o n_e}{\sqrt{n_e^2 \cos^2 \alpha + n_o^2 \sin^2 \alpha}} \quad (2.7)$$

where $\alpha = \theta$ in the present case where diffraction is neglected. Expressed as a function of θ , the free energy of the NLC is therefore

$$\begin{aligned} \mathcal{F} = \int_V d^3r \frac{1}{2} [& (\kappa_{11} \cos^2 \theta + \kappa_{33} \sin^2 \theta) (\partial_x \theta)^2 + (\kappa_{11} \sin^2 \theta + \kappa_{33} \cos^2 \theta) (\partial_z \theta)^2 + \\ & 2(\kappa_{33} - \kappa_{11}) \sin \theta \cos \theta \partial_x \theta \partial_z \theta - 2 S_z \frac{n_o}{c} \frac{1}{\sqrt{1-r \sin^2 \theta}}] \end{aligned} \quad (2.8)$$

where

$$r = 1 - \frac{n_o^2}{n_e^2},$$

is the dielectric anisotropy. It is a measure of the difference between ordinary and extraordinary indices of refraction. The steady-state angle distribution $\theta(x,z)$ is obtained by minimizing the free energy $\mathcal{F}$ subject to the boundary conditions $\theta(z=0) = \theta(z=d) = 0$. A variational calculation with respect to θ applied to the functional $\mathcal{F}$ gives the Euler-Lagrange equation

$$\frac{\partial \mathcal{F}}{\partial \theta} - \frac{\partial}{\partial x} \frac{\partial \mathcal{F}}{\partial (\partial_x \theta)} - \frac{\partial}{\partial z} \frac{\partial \mathcal{F}}{\partial (\partial_z \theta)} = 0 \quad (2.9)$$

which can be expressed in terms of θ as the director equation

$$(\kappa_{33} - \kappa_{11}) \sin \theta \cos \theta \left[(\partial_z \theta)^2 - (\partial_x \theta)^2 - 2 \frac{\partial^2 \theta}{\partial x \partial z} \right] - (\kappa_{33} - \kappa_{11}) (\cos^2 \theta - \sin^2 \theta) \partial_x \theta \partial_z \theta -$$

$$(\kappa_{11} \sin^2\theta + \kappa_{33} \cos^2\theta) \partial_{zz}^2 \theta - (\kappa_{11} \cos^2\theta + \kappa_{33} \sin^2\theta) \partial_{xx}^2 \theta - S_z \frac{n_0 r}{c} \frac{\sin\theta \cos\theta}{(1-r\sin^2\theta)^{3/2}} = 0. \quad (2.10)$$

This nonlinear partial differential equation is too complex to be solved analytically without further approximation. One often introduces the one elastic constant approximation, which neglects the anisotropy in the elastic constants and sets them all equal: $\kappa_{11} = \kappa_{22} = \kappa_{33} = \kappa$. This approximation simplifies the directors equation to

$$\partial_{xx}^2 \theta + \partial_{zz}^2 \theta + S_z \frac{n_0 r}{c\kappa} \frac{\sin\theta \cos\theta}{(1-r\sin^2\theta)^{3/2}} = 0. \quad (2.11)$$

In the case of small dielectric anisotropy or small reorientation angle, the denominator of the last term expressing the nonlinear coupling can be approximated by 1, reducing this equation to a sine-Gordon equation. The standard Freedericksz transition in a static magnetic field is described by a sine-Gordon equation of the same type. Its solutions have been discussed previously [see for example Ben-Abraham 1976]. They are in the form of the different modes of the elliptic sine function. For the more general case where r and θ are arbitrary, no analytical solution can be found and a numerical integration is necessary.

So far we have considered only the steady-state behaviour of the molecular reorientation. To describe the dynamics of the OFT, we must take into account the viscous properties of the liquid crystal medium. When the molecules are reoriented, a viscous torque tends to oppose rapid rotations of the directors. It limits the ability of the medium to respond instantaneously to a perturbation of its orientation by the laser field and thus governs the dynamics of the orientational effects induced by a laser beam. In the study of the dynamical OFT, a time dependent term must thus be introduced in the director equation to describe the dissipative effects due to the rotational viscosity γ of the NLC. This effect is modeled in the present work by the introduction of a phenomenological Debye type relaxation term in the directors equation. The time dependent distribution of the director vectors in the nematic sample obeys then the equation

$$\partial_{xx}^2 \theta + \partial_{zz}^2 \theta + S_z \frac{n_0 r}{c \kappa} \frac{\sin \theta \cos \theta}{(1 - r \sin^2 \theta)^{3/2}} = \frac{\gamma}{\kappa} \partial_t \theta. \quad (2.12)$$

This is the basic equation describing the dynamics of the optical Freedericksz transition in a nematic sample illuminated perpendicularly by a plane wave, linearly polarized, under the following assumptions;

- there is no diffraction of the beam,
- the absorption of light is negligible in the sample,
- the elastic anisotropy is neglected,
- the dynamics is governed by a Debye type relaxation of the directors.

II.3 DISCUSSION

Neglecting beam diffraction is justified, even past the plane wave approximation by the fact that the laser beam spot size ω_0 is typically of the same order of magnitude as the sample thickness d , which is chosen in the numerical examples of the order of 100 microns. The Fresnel number defined by [Svelto 1976]

$$f = \frac{\omega_0^2}{d \lambda}, \quad (2.13)$$

where λ is the wavelength of the light, is of the order of $f \simeq 10^3 \gg 1$ for typical parameters, which shows that diffraction does not play an important role and can be neglected.

The one elastic constant approximation does not allow to treat certain phenomena involving the elastic anisotropy, such as the intrinsic hysteresis which occurs in the nonlinear phase shift for certain values of the parameters when the ratios between the κ_{ij} are varied: Ong has shown that when different values of the elastic constants are permitted, the orientation of the molecules, and hence the phase shift of light can switch discontinuously from a zero to a non-zero value when the laser intensity reaches the threshold value. Decreasing the intensity again, it keeps a non-zero value for input intensities lower than the threshold (hysteresis). In this case, the Freedericksz transition is first-order [Ong 1983]. This situation is not allowed if

all elastic constants are taken to be equal. In that case the Freedericksz transition is always second-order. Still, in most of the cases, the Freedericksz transition is second-order and the one elastic constant approximation describes most experimental results with a good accuracy.

In the case where the transverse intensity profile of the laser is taken into account, the neglect of diffraction allows each radius x to be considered independently. The plane wave formulation of the electromagnetic term is then still valid for each x , and $S_z(x)$ remains conserved during propagation along z .

The model assumes no absorption of light and, as we just discussed, no coupling of different x . Thus, it neglects all the effects due to laser heating. This is a rather serious limitation: thermal properties of liquid crystals can be of considerable importance.

A large fraction of the liquid crystal media used in electro-optical devices are thermotropic: when the temperature T is varied, they undergo a first-order phase transition from a crystalline state to a liquid state, exhibiting in between one or more mesophases. In the simplest case, the isotropic crystalline phase is replaced by a liquid crystal mesophase when the temperature reaches a critical temperature T_1 . This phase remains stable up to a second critical temperature T_2 where a transition to the isotropic liquid phase occurs. At the transition between isotropic liquid and nematic mesophase, the index of refraction of the medium is discontinuous, jumping from the two characteristic values n_o and n_e of a birefringent medium to the isotropic value n_i of a liquid. These quantities are then temperature dependent over the whole range of T corresponding to the nematic phase; this dependence is schematically represented in Fig. 2.2.

In the presence of absorption, the laser induces a variation ΔT of the temperature in the sample, which in turn induces therefore a variation of the effective index of refraction, and thermally induced phase shifts of the light propagating through the liquid crystal. In the general case where laser heating in the medium is not negligible, the coupling between molecular reorientation and thermal diffusion leads to a complex analysis of the light induced effects in a NLC: The temperature increase ΔT must be determined considering the dependence of thermal diffusivity on the molecular reorientation, which in turn depends in a general case on ΔT through a temperature dependent index of refraction. Furthermore, heating can induce a convective motion of the molecules, coupling the molecular

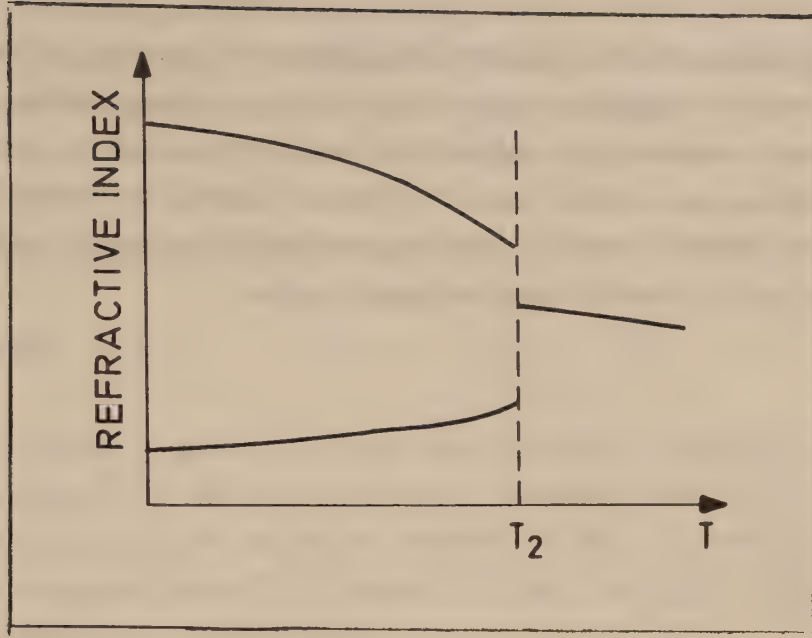


Fig. 2.2 Temperature dependence of the refractive indices of a liquid crystal. T_2 is the critical temperature for which the transition nematic mesophase $\rightarrow$ isotropic liquid occurs.

a hydrodynamical flow. This complex treatment can be avoided to some extent considering thin films of nematic liquid crystal, and by working at temperatures far from the nematic - isotropic transition. In this region, the variations of n_o and n_e with temperature are only of the order of $10^{-3}/^{\circ}\text{C}$ [Karat 1976]. The temperature variations of the medium parameters are then small, and their effects can be neglected when compared to those of molecular reorientation. The approximation is justified if we notice that the molecular reorientation is proportional to the dielectric anisotropy which is of the order of 10^{-1} .

The assumption concerning the dynamics of the OFT is important and simplifies the problem to a considerable measure. In the most general case, one must include the hydrodynamics of the liquid crystal, taking into account molecular motions. The director field $\mathbf{n}$ alone is then not sufficient to describe the optical properties of the NLC, but a velocity field $\mathbf{v}$ must be introduced, and coupled equations have to be solved for $\mathbf{n}$ and $\mathbf{v}$. In the geometry considered here, the reorientation due to the optical field does not induce any translational motion of the molecules. Their centers of gravity remain unperturbed, $\mathbf{v} = 0$ [De Gennes, 1975].

The director equation obtained using these different assumptions has to be solved for homeotropic boundary condition. Due to the complexity of the nonlinear term, an analytical solution can not be found in the general case, and one is reduced to a numerical treatment. But analytical solutions can be derived in certain limiting cases considering approximate forms of the director equation. We develop in the next Chapter two different analytical approaches, one valid in the case of small dielectric anisotropy, and the other for small reorientation regime.

III. TWO ANALYTICAL APPROXIMATIONS : SMALL DIELECTRIC ANISOTROPY AND SMALL REORIENTATION REGIME

III.1 SMALL DIELECTRIC ANISOTROPY

Spatial modes

In the case of a plane wave input laser propagating along the z-axis, the problem introduced in the preceding Section corresponds effectively to a one-dimensional situation, with translational invariance in the x dimension. At steady state, the reorientation angle θ is a function of z only, and S_z is a constant. The director equation (2.11) reduces then to

$$d_{zz}^2 \theta + \beta r \frac{\sin \theta \cos \theta}{(1 - r \sin^2 \theta)^{3/2}} = 0 \quad (3.1)$$

where $\beta = S_z n_0 / c\kappa$ is the scaled intensity of the input laser. A first integral of this equation can be performed, giving

$$\frac{1}{2} (d_z \theta)^2 + \frac{\beta}{(1 - r \sin^2 \theta)^{1/2}} = C, \quad (3.2)$$

where C is an integration constant.

In the case of a small dielectric anisotropy, characterized by $r \ll 1$ (that is : $n_o \cong n_e$, this situation corresponds to a small birefringence), an expansion to first order in r of the square root leads to

$$\frac{1}{2} (d_z \theta)^2 + \beta \left[1 + \frac{r}{2} \sin^2 \theta \right] = C. \quad (3.3)$$

The integration constant C may be expressed in terms of the extremum reorientation angle θ_M , for which $d_z \theta = 0$

$$C = \beta \left[1 + \frac{r}{2} \sin^2 \theta_M \right] \quad (3.4)$$

A formal integration yields an implicit expression for θ in terms of the elliptic integral

$$\int_0^{\theta(z)} \frac{d\theta'}{\sqrt{1 - \mu^2 \sin^2 \theta'}} = \pm \sqrt{\beta r} \frac{1}{\mu} z \quad (3.5)$$

where $\mu^2 = 1/\sin^2 \theta_M$.

The solution of this integral equation is an elliptic sine function, leading to the following expression for θ

$$\theta(z) = \pm \sin^{-1} \left[\frac{1}{\mu} \operatorname{sn} \left[\sqrt{\beta r} z, \frac{1}{\mu} \right] \right] . \quad (3.6)$$

The elliptic sine function $\operatorname{sn}(\xi, 1/\mu)$ is periodic of period $4K(1/\mu)$, where K is the complete elliptic integral

$$K \left[\frac{1}{\mu} \right] = \int_0^{\frac{\pi}{2}} \frac{d\theta}{\sqrt{1 - 1/\mu^2 \sin^2 \theta}} . \quad (3.7)$$

The elliptic sine $\operatorname{sn}(\xi, 1/\mu)$ has an infinite number of zeros at $\xi = 2mK(1/\mu)$, m being an integer [Byrd and Friedman 1954]. Using these general properties of the sn -function, combined with the homeotropic boundary condition $\theta(z=d) = 0$, we obtain different possible solutions for $\theta(z)$. (The first boundary condition $\theta(z=0) = 0$ is automatically satisfied since $z = 0$ is always a zero of the elliptic sine.) Setting $\sqrt{\beta r} d$ equal to each successive zero of the elliptic sine, the different solutions can all be expressed by the form (3.6), but are characterized by different values of μ corresponding to the successive values of m . We call these solutions "spatial modes" of the director angle. For each m , the corresponding value of the modulus $1/\mu$ is

obtained as the solution of the equation

$$\operatorname{sn}\left[\sqrt{\beta r} \frac{d}{2m}, \frac{1}{\mu}\right] = 1. \quad (3.8)$$

Using the complete elliptic integral $K(1/\mu)$ defined in (3.7), the solution of Eq.(3.8) is expressed as

$$\sqrt{\beta r} \frac{d}{2m} = K\left[\frac{1}{\mu}\right]. \quad (3.9)$$

K is a monotonically growing function of its argument, with $K(0) = \pi/2$, which implies that each mode m only becomes possible for intensities larger than the threshold value

$$\beta_{\text{th}}^m = \frac{\pi^2 m^2}{d^2 r}, \quad (3.10)$$

as illustrated in Fig. 3.1.

The lowest threshold intensity is that for the fundamental mode $m = 1$. It corresponds to the "conventional" threshold of the OFT [Zel'dovich and Tabyiryan 1982, Ong 1983].

$$\beta_{\text{th}} = \frac{\pi^2}{d^2 r}. \quad (3.11)$$

For intensities smaller than β_{th} , the only possible solution for the reorientation angle is $\theta(z) = 0$ for all z , corresponding to the directors aligned on the easy direction. The laser intensity is not strong enough to overcome the elastic forces, there is no molecular reorientation. As the intensity β is increased, the fundamental mode $m = 1$ becomes first possible and a nonhomogenous θ characterizes the molecular reorientation distribution. For even higher intensities, the higher order spatial modes can be reached. Fig. 3.2 illustrates the first and the second of these modes, the first one being plotted for two values of the laser intensity β .

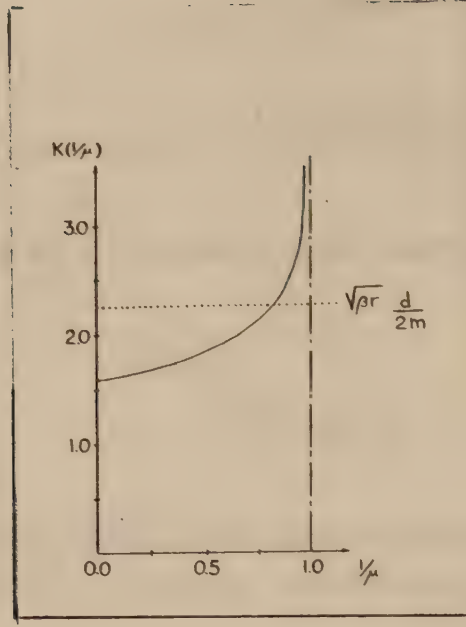


Fig. 3.1 Complete elliptic function $K(1/\mu)$. The point where it intersects the straight line $\sqrt{\beta r} d/2m$ determines the modulus μ .

In all the numerical calculations presented in this work, the liquid crystal parameters correspond to MBBA (methoxybenzylidene-butylaniline). This medium is the "sodium" of liquid crystals, it is very often used in experiments and its parameters are well known: $n_o = 1.54$, $n_e = 1.75$, $\kappa = 7.5 \cdot 10^{-12} \text{N}$ and $\gamma = 0.8 \text{P}$.

As seen from Eq.(3.11), the threshold intensity of the OFT is inversely proportional to the square of the sample thickness (this is similar to the threshold field H_c in the case of a Freedericksz transition in a static magnetic field, where H_c is proportional to π^2/d^2 [De Gennes 1975]). In the fundamental mode, the maximum reorientation angle is reached in the middle of the sample, where the fixed anchoring at the surfaces has the weakest influence.

The threshold for higher spatial modes increases quadratically with the order of the mode. Such an increase was to be expected intuitively, since for higher modes, $\theta(z)$ varies on a shorter scale in the sample; the distortions induce then a higher elastic energy, which can only be overcome by a higher electromagnetic energy. Furthermore, higher modes are characterized by the extrema of reorientation

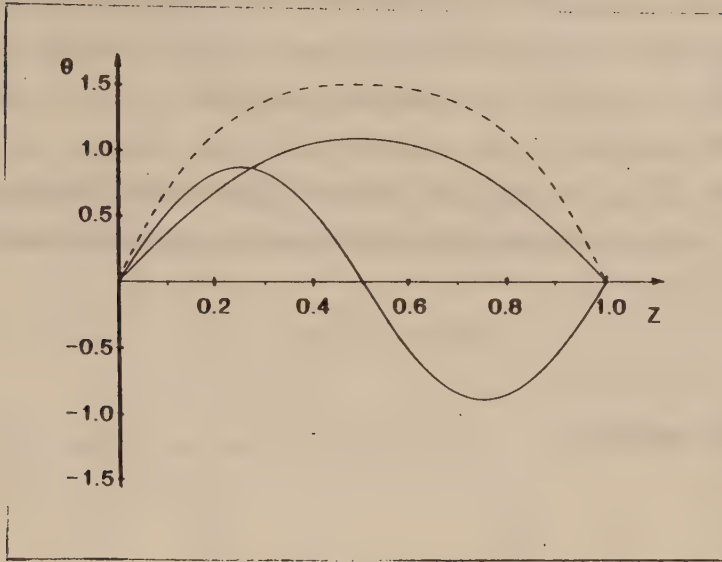


Fig. 3.2 Reorientation angle distribution $\theta(z)$ for the first and second order spatial modes (z in units of d). For the 1st mode, $\beta = 2$ (solid line) and $\beta = 6$ (dashed line). For the 2nd mode, $\beta = 6$. In all cases, as well as in the following figures, β is scaled to β_{th} . The value of the dielectric anisotropy is fixed to $r = .23$, corresponding to $n_o = 1.54$ and $n_e = 1.75$. The elastic constant is set equal to the typical value $\kappa = 7.5 \cdot 10^{-12} \text{N}$.

occurring closer to the boundaries, where the molecules are strongly anchored. For this reason, the maximum value of the reorientation decreases with m , as can be seen from Fig. 3.1.

For high enough intensities, the maximum reorientation angle is $\pi/2$. It corresponds to the molecules totally aligned with the field and at right angle to the easy direction. Since the molecules are symmetric and the orientations $\mathbf{n}$ and $-\mathbf{n}$ are not distinguishable, all possible configurations of the director orientations can be described by a continuous function $\theta(z)$ bounded by $\pi/2$. When the input intensity is increased from the threshold value, the maximum value of $\theta(z)$ increases until it reaches $\pi/2$ at the extrema; if the input intensity is further increased, the spatial domain where $\theta(z) = \pi/2$ becomes larger around the extrema, and a saturation occurs. This is illustrated in Fig. 3.2, where $\theta(z)$ is shown in the fundamental mode for two different intensities.

We note that in the case of a standard Freedericksz transition induced by a static magnetic field, the sine-Gordon equation (3.3) describes the molecular reorientation for a general case, without restriction to small values of the parameters. The solutions of this problem have been discussed in terms of the different modes of the elliptic sine function. Each mode becomes successively possible when the static magnetic field is increased past the corresponding threshold. [Ben Abraham 1976].

Linear stability analysis

The spatial modes (3.6) are all steady-state solutions of the director equation. The fundamental mode is the easiest to reach, since it involves a lower elastic energy. The higher order modes are possible solutions, but they correspond to larger input intensities and seem intuitively less favorable for the system, since they involve higher energies. Which solutions are stable can however not be described by a steady-state treatment. In a real system, unavoidable small perturbations will lead to a decay of unstable steady-state solutions.

To study the stability of the steady-state solutions against small perturbations, a linear stability analysis is performed, using the expansion to first order in r of the equation of motion (2.12) obtained in Chapter II, restricting it to only one spatial dimension z ,

$$\partial_{zz}^2 \theta + \beta r \sin\theta \cos\theta = \frac{\gamma}{\kappa} \partial_t \theta \quad (3.12)$$

with $\theta(z,t)$ being now a function of both position and time.

To perform a linear stability analysis of a given steady-state solution θ_{st} , we perturb it by a small time dependent quantity ϵ and linearize the resulting equation of motion in ϵ . If the perturbation decays with time, the solution evolves back towards the initial steady-state solution, θ_{st} is stable. On the other hand, if the perturbation grows with time, θ_{st} is an unstable solution.

Specifically, we proceed by expressing $\theta(z,t)$ as

$$\theta(z,t) = \theta_{st}(z) + \epsilon(z,t) . \quad (3.13)$$

Introducing this expression into Eq.(3.12), factorizing $\epsilon(z,t)$ as $A(z)e^{\Gamma t}$ and keeping only terms linear in ϵ yields the eigenvalue equation

$$d_{zz}^2 A + A[g(z) - \lambda] = 0 , \quad (3.14)$$

where $\lambda = \Gamma\gamma/\kappa$ and the potential $g(z)$ is given by

$$g(z) = \beta r \left[1 - \frac{2}{\mu^2} \operatorname{sn}^2 \left[\sqrt{\beta r} z, 1/\mu \right] \right] . \quad (3.15)$$

The stability of $\theta_{st}(z)$ against small perturbations is determined by the sign of the maximum eigenvalue λ_M , which must be negative for the steady state to be stable.

We chose to determine the eigenvalues λ by expanding the eigenfunctions A on an orthogonal basis of $\{\sin(n\pi z/d)\}$, n integer

$$A = \sum_n a_n \sin(n\pi z/d) . \quad (3.16)$$

Using this expansion, multiplying both sides of (3.14) by $\sin(m\pi z/d)$ and integrating over z leads to

$$\sum_n a_n L_{nm} = \lambda_m a_m \quad (3.17)$$

where

$$L_{nm} = \frac{2}{d} \int_0^d dz \sin(n\pi z/d) [d_{zz}^2 + g(z)] \sin(m\pi z/d) \quad (3.18)$$

The eigenvalues λ_m are obtained by a numerical diagonalization of L . The set of $\{\sin(n\pi z/d)\}$ used to compute the eigenvalues is finite, but it is chosen large enough so that λ_M converges to a value which remains constant when the number of basis functions used is further increased. (We found that it was usually sufficient to expand A on a basis of 32 sine functions.)

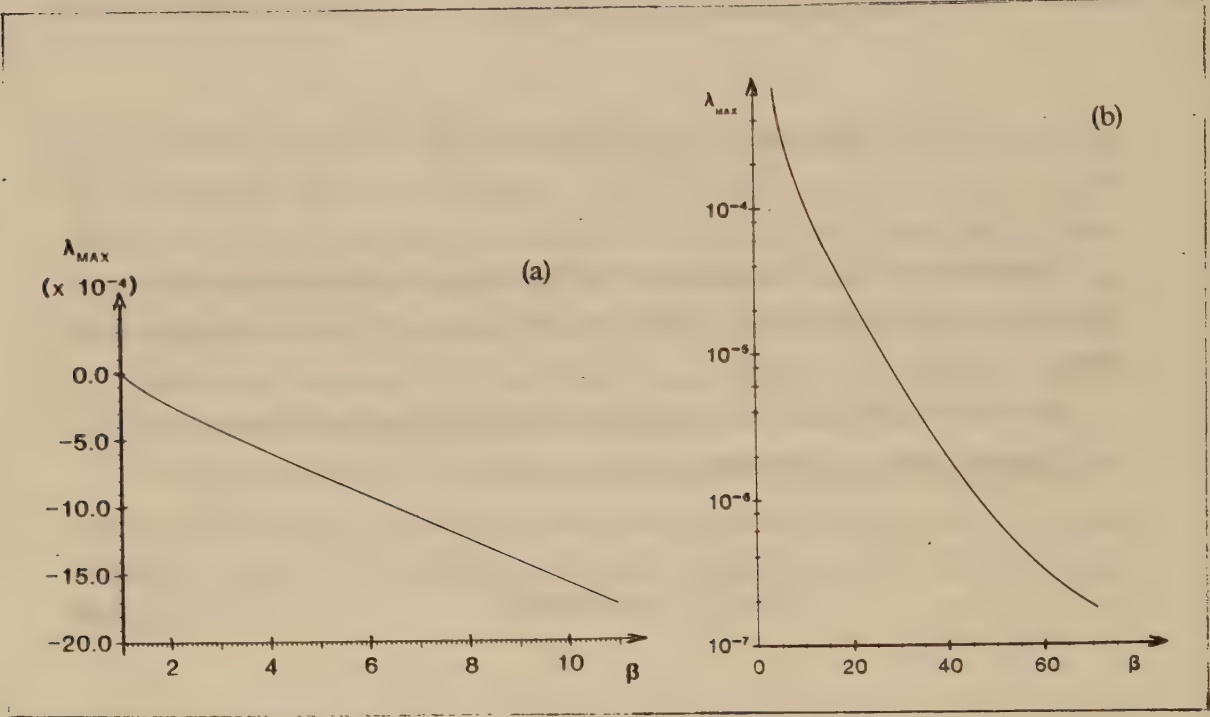


Fig. 3.3 Maximum eigenvalue λ_M as a function of β , for (a) the first and (b) the second sn-mode. The eigenvalues are calculated using 8 sine functions in the first case and 32 in the second. In both cases, a further increase of the number of basis functions leave the eigenvalues unchanged. The parameters are characteristic of MBBA, $d = 250\mu\text{m}$.

The results of this analysis are presented in Fig. 3.3 for parameters characteristic of a MBBA sample of length $d = 250 \mu\text{m}$. The maximum eigenvalue

λ_M is plotted as a function of β for (a) the fundamental mode, and (b) the second spatial mode $m = 2$. In the first case, λ_M is a monotonically decreasing function of β , from the value $\lambda_M = 0$ at threshold. This indicates that the first-order mode is stable against small perturbations. In the second case, in contrast, λ_M is always positive, the mode $m = 2$ is always unstable. It decreases monotonically with β to asymptotically reach 0 as $\beta \rightarrow \infty$.

For the higher modes, the number of sine functions needed to obtain reliable results is increased, making a complete study of λ_M for a large β -domain expensive in computer time. But the points that were checked lead with a high degree of confidence to the conclusion that the higher spatial modes are unstable.

Dynamical solution

Before proceeding with a fully numerical study of the dynamics of the NLC, it is instructive to first consider the dynamical behaviour of θ close to threshold. In this case, the reorientation angle is very small and a linearized analysis can be performed. Linearizing the dynamical equation (3.12) in θ allows to determine analytically the time constant τ characterizing the onset of the fundamental mode. We assume a separable form of θ , seeking a solution of the form

$$\theta(z,t) = \zeta(z)e^{t/\tau} , \quad (3.19)$$

the threshold value being then obtained by requiring that τ becomes positive. Past this value, θ grows exponentially with time. The calculation leads to the following expression for τ

$$\tau_{\text{on}} = \left[\frac{\kappa}{\gamma} \frac{\pi^2}{d^2} \frac{\beta - \beta_{\text{th}}}{\beta_{\text{th}}} \right]^{-1} . \quad (3.20)$$

This dependence describes critical slowing down and is characteristic of second-order phase transition.

The predictions of the linear stability analysis performed in the preceding Section were confirmed by a numerical solution of the dynamical problem (2.12). The time-dependent solutions are computed using the general one-dimensional equation valid for all values of r , with a small numerical value of the parameter r ($r = .23$ for MBBA, the nematic chosen for the numerical applications). The fundamental mode is effectively stable. It is reached for an input intensity larger than β_{th} , with $\theta(z,t=0) = \epsilon$ as initial condition. Fig. 3.4 illustrates the numerical results. Cases (a) and (b) correspond to two different values of the parameter ϵ , set equal respectively to 10^{-5} and 10^{-2} , with an input intensity $\beta = 2 \beta_{th}$ in both cases. ϵ corresponds to a uniform small "fluctuation" necessary for the system to leave the unstable state. The time for these "fluctuations" ϵ to drive the system out of the unstable uniform state is shorter for larger values of ϵ . This will be discussed later when thermal fluctuations are considered in a stochastic model. Case (c) displays the time evolution for a larger input intensity equal to $\beta = 10 \beta_{th}$ for $\epsilon = 10^{-2}$. It is noticeable that even if the input intensity is large enough to induce in principle a second or even a third mode of the reorientation, the system evolves towards the fundamental angle distribution. Furthermore, when compared to the evolution displayed in (b), case (c) shows that for a given ϵ , the evolution towards the steady-state is faster for a larger intensity, as it is expected from critical slowing down occurring close to threshold, expressed by Eq.(3.20).

Fig. 3.5 illustrates a situation where the initial condition $\theta(z,t=0)$ is chosen close to the second spatial mode, $\theta(z,t=0) = \sin(2\pi z/d)$, and the input intensity larger than β_{th}^2 . In that case, the system first evolves towards the second mode and is trapped for some time in this configuration before finally collapsing to the fundamental mode. Figure (a) corresponds to an input intensity of $6 \beta_{th}$. In this case, the system is trapped in the vicinity of the second mode for more than 400 seconds before escaping this state and evolving towards the stable mode $m = 1$. Case (b) displays the evolution of the same system, but for an input intensity of $15 \beta_{th}$. Here the second sn-mode appears to be stable for time up to 4×10^3 seconds, before the system again collapses to the fundamental mode. The results of the linear stability analysis confirm this behaviour: the decrease of λ_M with β shows that a small perturbation of the second mode will grow faster when β is closer to the threshold than when β is larger, see Fig.3.3(a).

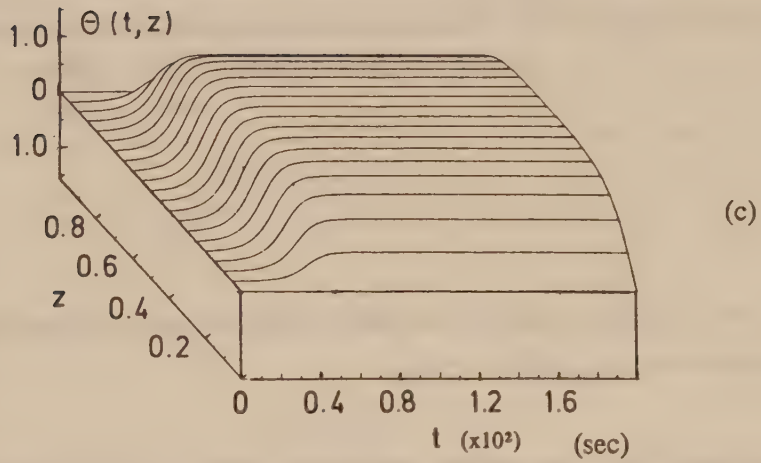
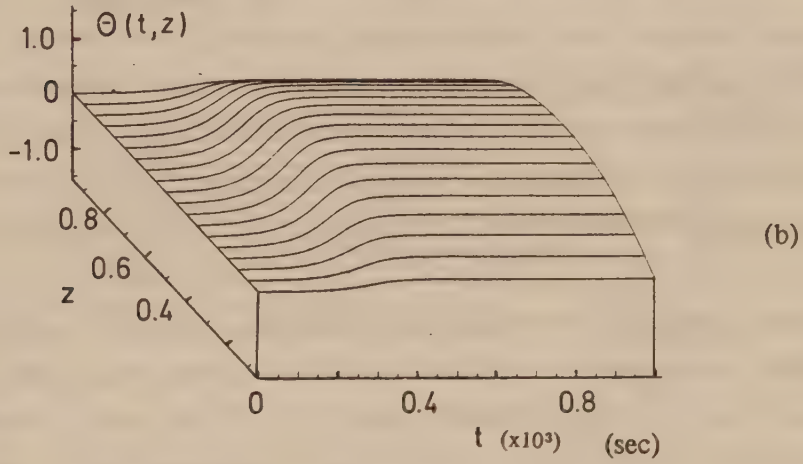
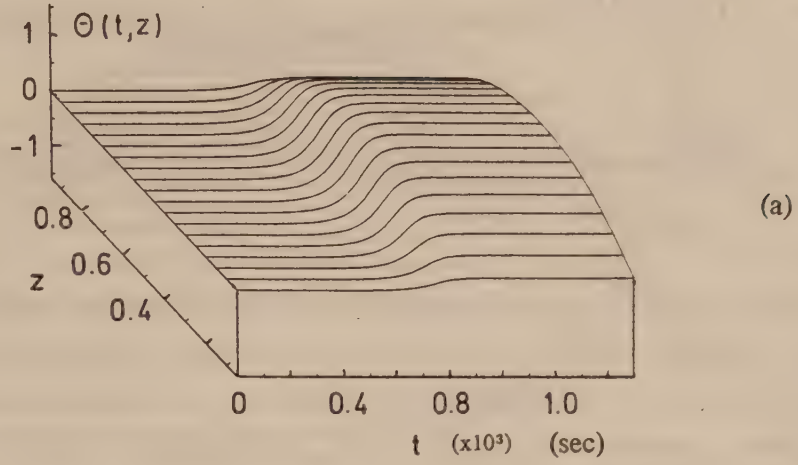


Fig. 3.4 Evolution of the molecular reorientation from the initial condition $\theta(z,t=0) = \epsilon$, for $d = 250 \mu\text{m}$. (a) $\epsilon = 10^{-5}$, $\beta = 2 \beta_{\text{th}}$; (b) $\epsilon = 10^{-2}$, $\beta = 2 \beta_{\text{th}}$; (c) $\epsilon = 10^{-2}$, $\beta = 10 \beta_{\text{th}}$.

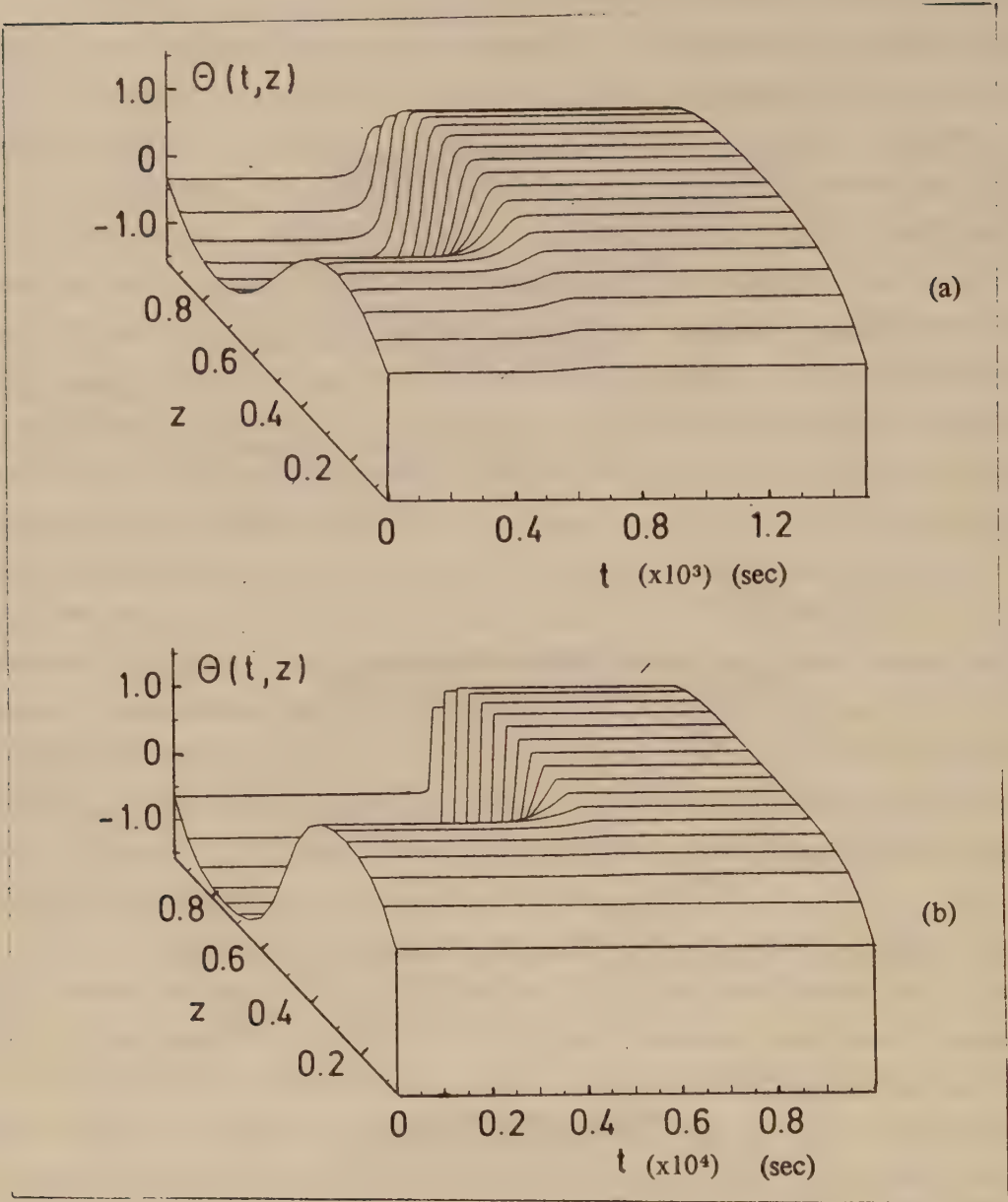


Fig. 3.5 Time evolution from the second to the first mode, for two different input intensities. (a) $\beta = 6 \beta_{th}$; (b) $\beta = 15 \beta_{th}$.

In summary, the time during which the system can be trapped in a higher unstable mode can be extremely long, but within the present model there is no way to reach these higher modes continuously by increasing β from $\beta < \beta_{th}$. We will see

in Chapter V that when thermal fluctuations are taken into account, higher modes of the molecular reorientation can be naturally excited simply by inducing a Freedericksz transition with a laser of large enough intensity.

III.2 TWO DIMENSIONAL MODEL: SMALL REORIENTATION REGIME

The approximate solution derived in the preceding Section is useful to study in a simplified one-dimensional situation the static and dynamical aspects of the longitudinal modes describing the reorientation of the directors in an OFT. A one-dimensional model of the OFT, however, completely neglects transverse correlations of the molecular reorientation, due to the elastic forces between the molecules. This effect can not be neglected if the laser beam waist ω_0 is of the same order or smaller than the sample thickness d . We see in this Section that transverse diffusion can lead to inhomogeneities in the transverse profile of the reorientation even under plane wave pumping. A two-dimensional model is considered now to take these effects into account and discuss the transverse distribution of the molecular reorientation.

In this Section, we consider a simplified two-dimensional model and specifically restrict our analysis to the case of small reorientations, corresponding to intensities close to threshold. In this limit, Eq.(2.11), which describes the steady-state molecular reorientation due to laser illumination is amenable to a partial analytical treatment.

For small angles θ , the nonlinearity describing the electromagnetic coupling can be developed in a power series, which gives to third order

$$\frac{\sin\theta\cos\theta}{(1-r\sin^2\theta)^{3/2}} \cong \theta - \alpha\theta^3 \quad (3.21)$$

with

$$\alpha = \frac{2}{3} - \frac{3r}{2} . \quad (3.22)$$

In this limit, Eq.(2.11) can be interpreted as the Euler-Lagrange equation resulting from the lagrangian density

$$2L = (\partial_x \theta)^2 + (\partial_z \theta)^2 - \beta r(\theta^2 - \alpha \theta^4/2) \quad (3.23)$$

with the variational principle $\delta \mathcal{L} = 0$, where

$$\mathcal{L} = \int dx \int dz L . \quad (3.24)$$

The homeotropic boundary conditions $\theta(z=0) = \theta(z=d) = 0$ suggest expanding the solution $\theta(x,z)$ in a Fourier series

$$\theta(x,z) = \sum_{n=1}^{\infty} a_n(x) \sin(n\pi z/d) . \quad (3.25)$$

For intensities close to threshold, the only possible longitudinal mode is the fundamental one, as was shown in the preceding Section. The higher modes require higher incident intensities, which are outside the domain of validity of the small reorientation approximation. The sum (3.25) then can be restricted to its first term

$$\theta(x,z) = a(x) \sin(\pi z/d) . \quad (3.26)$$

With this form of the solution, the lagrangian (3.24) can be integrated over z , and a variational calculation leads to an equation for the transverse dependence $a(x)$ of the reorientation angle θ only :

$$d_{xx}^2 a + [r\beta(x)(1 - 3\alpha a^2/4) - \pi^2/d^2] a = 0 . \quad (3.27)$$

The intensity profile β of the pump beam is now a function of x . In the simple case of a plane wave input intensity, β is constant and (3.27) becomes

$$d_{xx}^2 a + a(\sigma - \xi a^2) = 0 , \quad (3.28)$$

where

$$\sigma = \beta r - \pi^2/d^2$$

and

$$\xi = 3\alpha\beta r/4 . \quad (3.29)$$

We restrict our discussion to positive values of ξ , implying that the dielectric anisotropy is restricted to the domain $0 < r < 4/9$. The homogeneous solutions of (3.28) are easily found by dropping the second-order derivative in Eq. (3.28). For $\sigma > 0$ we readily find the solutions

$$a = 0$$

and

$$a_{\infty} = \pm\sqrt{\sigma/\xi} , \quad (3.30)$$

whereas for $\sigma < 0$ only the zero solution applies. $\sigma = 0$ corresponds to $\beta = \pi^2/d^2r$, which is precisely the threshold intensity obtained in the small dielectric anisotropy case (Eq.3.11). The new solutions appearing as the intensity is increased past $\sigma = 0$ simply reflect the fact that the director angle exhibits a second-order phase transition at the OFT threshold. In analogy to mechanics, one can formally introduce a potential V , the transition is then associated to the appearance of new minima of V as the order parameter σ is increased. The potential corresponding to Eq.(3.28) is $V = -\sigma a^2/2 + \xi a^4/4$. For $\sigma < 0$, V has a single minimum at $a = 0$. For $\sigma > 0$, $a = 0$ becomes a local maximum, and new minima appear at $a = \pm a_{\infty}$, as illustrated in Fig. 3.6.

The curvature of the potential at the extrema is a direct criterion of the stability of each solution. $a = 0$ is stable for $\sigma < 0$ and unstable above threshold. $a = \pm a_{\infty}$ is a stable solution for $\sigma > 0$.¹

¹ The case $\xi < 0$ is characterized by a stable constant solution $a = 0$ for $\sigma < 0$, which becomes metastable at $\sigma = 0$. A third-order expansion is not sufficient to obtain stable solutions past this point, and the next (5th) order must be included. This case is not considered here, where the restriction $\xi > 0$, $0 < r < 4/9$ applies.

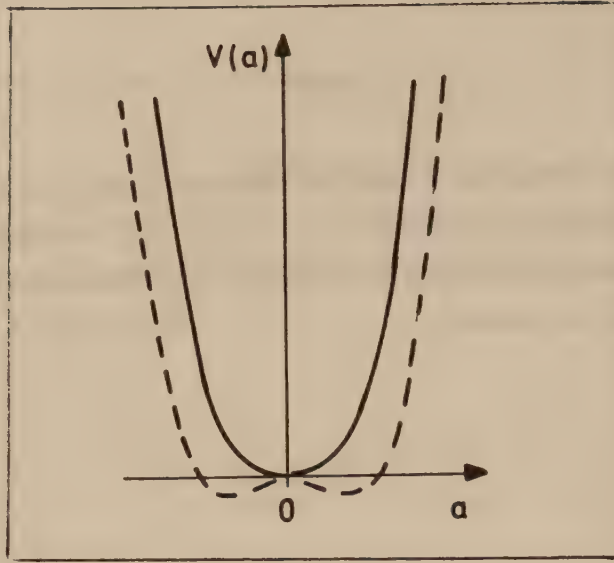


Fig. 3.6 Potential V as a function of a , for arbitrary units. The full curve corresponds to $\sigma < 0$ and represents the situation under threshold; the dashed curve corresponds to $\sigma > 0$ and displays the situation above threshold.

The inhomogenous solutions of Eq.(3.28) are shown in Appendix B to be

$$a(x) = a_{\infty} \tanh \left[a_{\infty} \sqrt{\xi/2} x + C_1 \right] \quad (3.31)$$

and

$$a(x) = a_{\infty} \coth \left[a_{\infty} \sqrt{\xi/2} x + C_2 \right] , \quad (3.32)$$

where C_1 and C_2 are arbitrary constants of integration. The solution (3.32) is clearly nonphysical.

Eq. (3.31) indicates that even under plane wave illumination, the transverse coupling between the molecules of the liquid crystal induces a transverse anisotropy in the reorientation angle θ for sufficiently high incident intensities. The location at

which switching between the solutions a_{∞} and $-a_{\infty}$ takes place along the x -axis is however undetermined.

Above threshold, four possible behaviours of the reorientation are thus possible: 3 homogeneous ones, $a = a_{\infty}$, 0 , $-a_{\infty}$, and the inhomogeneous hyperbolic tangent solution.

Stability analysis

The stability of these steady-state solutions is studied analytically by expanding the nonlinearity in Eq.(2.12) to third-order in θ and seeking a solution of the type $\theta(x,z,t) = a(x,t) \sin(\pi z/d)$. The resulting equation of motion is the nonlinear diffusion equation

$$\frac{\gamma}{\kappa} \partial_t a(x,t) - \partial_{xx}^2 a(x,t) = [\beta r - \pi^2/d^2] a(x,t) - 3/4 \alpha \beta r a^3(x,t) \quad . \quad (3.33)$$

Each steady-state solution is perturbed by a small quantity ϵ

$$a(x,t) = a_{st}(x) + \epsilon(x,t) \quad . \quad (3.34)$$

Factorizing $\epsilon(x,t)$ as $A(x)\exp(\Gamma t)$ and introducing this form in Eq.(3.33) yields

$$d_{xx}^2 A + [\beta r - \pi^2/d^2 - 9/4 \alpha \beta r a_{st}^2 - \gamma \Gamma/\kappa] A = 0 \quad . \quad (3.35)$$

The stability of a_{st} is given by the sign of Γ , which is negative if a_{st} is stable against small perturbation.

The stability of the homogenous solutions is obtained by dropping the spatial derivative in (3.35), i.e. by

$$\beta r - \pi^2/d^2 - 9\alpha\beta r/4 a_{st}^2 < 0 \quad . \quad (3.36)$$

The zero solution $a_{st} = 0$ is stable for $\beta < \beta_{th}$ and is unstable if $\beta > \beta_{th}$, as can be deduced directly from (3.36). In this case, Γ is given by $[\beta r - \pi^2/d^2]\kappa/\gamma$, which is

negative below and becomes positive above threshold. For the stability of the nonzero homogenous solutions, (3.36) leads to $\Gamma = -2[\beta r - \pi^2/d^2]\kappa/\gamma$, which is always negative for β above threshold. The homogenous nonzero solutions are always stable, as expected from the mechanical potential analogy.

The stability of the inhomogenous steady-state solution (3.31) is obtained by substituting its value into (3.35). Introducing

$$y = x + C_1/\nu$$

$$\nu = \sqrt{1/2 (\beta r - \pi^2/d^2)}$$

$$\mu = 3 (\beta r - \pi^2/d^2)$$

$$\Omega = -2 (\beta r - \pi^2/d^2) - \gamma/\kappa \Gamma \quad ; \quad (3.37)$$

yields the eigenvalue equation

$$d_{yy}^2 A(y) + \left[\Omega + \frac{\mu}{ch^2(\nu y)} \right] A(y) = 0 \quad . \quad (3.38)$$

Its eigenvalues are given by [Landau and Lifshitz 1977]

$$\Omega_n = -\nu^2/4 \left[-(1+2n) + \sqrt{1 + 4\mu/\nu^2} \right]^2 \quad , \quad (3.39)$$

and must satisfy the condition

$$\Omega_n \geq -\mu \quad . \quad (3.40)$$

This implies that

$$\Gamma_n = -\kappa/2\gamma (\beta r - \pi^2/d^2)n(4 - n) \quad , \quad (3.41)$$

together with the condition

$$\Gamma_n \leq \kappa/\gamma (\beta r - \pi^2/d^2) , \quad (3.42)$$

which limits the possible values of n to $n \leq 4$. For these values, Γ_n is always negative for $\beta \geq \beta_{th}$, that is, the inhomogeneous steady-state solution is stable. This demonstrates the possibility of inducing a stable, inhomogeneous transverse distribution of directors under homogeneous plane-wave pumping.

The dynamics of the reorientation is described in the small reorientation regime by Eq.(3.33). Such nonlinear diffusion equations have been studied in considerable detail in biology, where they describe selection-migration processes. They are known [Fife 1970, Albano et al. 1984] to sustain traveling-wave solutions of the form $a(x,t) = a(x - vt)$. In the specific case considered here, 3 generic types of wavefronts are obtained, all others being deduced by translation and parity inversion. Taking a_∞ with the + sign in (3.30), they can be expressed as

$$a_{12}(x,t) = \frac{1}{2} a_\infty \left\{ -1 + \tanh \left[\frac{\rho a_\infty}{4} \left(x + \frac{3}{2} \frac{\kappa}{\gamma} \rho a_\infty t \right) \right] \right\} ,$$

$$a_{13}(x,t) = a_\infty \tanh \left[\sqrt{\frac{3}{8}} \alpha \beta r a_\infty x \right] ,$$

$$a_{23}(x,t) = \frac{1}{2} a_\infty \left\{ 1 + \tanh \left[\frac{\rho a_\infty}{4} \left(x + \frac{3}{2} \frac{\kappa}{\gamma} \rho a_\infty t \right) \right] \right\} . \quad (3.43)$$

where $\rho = \sqrt{3\alpha\beta r/2}$.

The wave front a_{13} that links the two stable solutions $\pm a_\infty$ is stationary. The other two, which connect the unstable solution $a = 0$ to one of the stable ones, propagate at the intensity-dependent velocity

$$v = \frac{3}{2} \frac{\kappa}{\gamma} \rho a_\infty , \quad (3.44)$$

to bring the whole crystal to a stable homogeneous configuration. It has been conjectured [Fife 1970] that the traveling-wave solutions (3.43) are the only stable solutions of the nonlinear diffusion equation of the type (3.33).

III.3 EFFECT OF THE FINITE SPOT SIZE OF THE LASER BEAM

The discussion presented in the first part of this Chapter considers plane wave input beams. In a real experiment, the laser beam has a transverse profile and a finite spot size; this can modify the characteristic parameters describing the OFT, such as the threshold value of the intensity. This Section gives an approximate analytical description of these effects in the small reorientation regime.

The transverse profile of the incident beam is taken for simplicity as a quadratic function:

$$\begin{aligned} \beta(x) &= \beta(1 - 4x^2/\omega_0^2) \quad \text{for } |x| \leq \omega_0/2 \\ &= 0 \quad \text{otherwise} \end{aligned} \quad (3.45)$$

We limit ourselves to the small reorientation regime and assume that $\theta(x,z)$ is of the form (3.26). The symmetry of the pump intensity about $x = 0$ suggests for $|x| \leq \omega_0/2$ an expansion of $a(x)$ as

$$a(x) = a_0 - a_2x^2 + a_4x^4 - \dots \quad (3.46)$$

We assume that $a_2 \ll a_0$ and keep only terms up to second power in x . This is consistent near the threshold of the Freedericksz transition. Inserting this Ansatz into Eq. (3.27) yields

$$a_2 = a_0r(\beta - \pi^2/d^2r - 3\beta\alpha a_0^2/4)/2 \quad (3.47)$$

For $|x| \geq \omega_0/2$, the solution of Eq.(3.27) is the exponential

$$a(x) = C \exp(-\pi|x|/d) \quad (3.48)$$

where C is a constant of integration to be determined.

At $|x| = \omega_0/2$, the solution (3.46) and its first derivative must go continuously into the solution (3.48). These two conditions allow to determine the two constants a_0 and C . Three solutions are possible :

$$a_0 = 0 \quad (3.49a)$$

as well as

$$a_0 = \pm \sqrt{\frac{(\beta - \pi^2/d^2r)(\omega_0/2 + \pi\omega_0^2/8d) - \pi/dr}{3\beta\alpha\omega_0(1 + \pi\omega_0/4d)/8}}, \quad (3.49b)$$

with

$$C = a_0 e^{\pi\omega_0/2d} \left[1 - \frac{\omega_0^2 r}{8} \left(\beta - \pi^2/d^2r - \frac{3}{4} \beta\alpha a_0^2 \right) \right]. \quad (3.50)$$

The zero solution is always possible, but the nonzero solutions for a_0 are possible only if

$$\beta \geq \frac{\pi^2}{d^2r} + \frac{8\pi}{\omega_0 r} (4d + \pi\omega_0)^{-1}. \quad (3.51)$$

The first term on the right-hand side of Eq. (3.51) is the plane wave OFT threshold intensity (3.11). For a positive dielectric anisotropy, the finite spot size increases the threshold compared to this plane wave value. It is expected that the smaller the radius of the incident beam, the higher the threshold intensity of the OFT [Csillag et al. 1982], and this is indeed the prediction of expression (3.51).

The general behaviour of the threshold intensity as a function of the beam waist is confirmed by a simple second calculation considering a transverse intensity profile of the form $\beta(x) = \beta_0 \text{sech}^2(x/\omega_0)$. The threshold can be determined in a linearized calculation considering only terms proportional to $a(x,t)$ in the dynamical equation (3.33), and neglecting the third-order contribution

$$\frac{\gamma}{\kappa} \partial_t a(x,t) - \partial_{xx}^2 a(x,t) = [\beta_0 r \operatorname{sech}^2(x/\omega_0) - \pi^2/d^2] a(x,t) , \quad (3.52)$$

valid near threshold. We seek a solution of the form $a(x,t) = f(x)\exp(t/\tau)$ and obtain the threshold value of β by requiring that τ becomes positive. This gives

$$\beta_0 = \frac{\pi^2}{d^2 r} + \frac{\pi}{dr\omega_0} . \quad (3.53)$$

The form (3.53) of the threshold implies again that the threshold is larger than the plane wave threshold β_{th} when the finite beam waist is considered, and is a decreasing function of ω_0 , reaching the value β_{th} in the limit $\omega_0 \rightarrow \infty$. The thresholds for these two different profiles are illustrated as functions of the laser beam waist in the next Chapter, (Fig. 4.3), and compared to the result of a numerical determination of the threshold.

IV. NUMERICAL SOLUTION OF THE DIRECTORS EQUATION

IV.1. STEADY STATE

The two models described in the preceding Chapter are limited to either small dielectric anisotropy or to the small reorientation regime. In the general case where the nonlinearity of the liquid crystal is considered to all orders in r and θ , an analytical solution can not be found and a numerical integration of the equations is necessary. This Chapter presents the method used to study numerically the OFT in a NLC and discusses the main results of this numerical experiment.

We consider the two-dimensional problem of a nematic sample of length d and infinite transverse dimension x , illuminated by a laser beam. The intensity profile of the input beam is taken to be gaussian of peak intensity β_0 and beam waist ω_0 . As before, diffraction and absorption are neglected and all elastic constants of the liquid crystal are taken equal to κ . We first study the system in steady state. In this case θ verifies the time-independent version of the second-order partial differential equation (2.12) in x and z

$$\frac{\partial^2}{\partial x^2} \theta + \frac{\partial^2}{\partial z^2} \theta + \beta(x)r \frac{\sin\theta\cos\theta}{(1-r\sin^2\theta)^{3/2}} = 0 , \quad (4.1)$$

where

$$\beta(x) = \beta_0 \exp(-x^2/\omega_0^2) . \quad (4.2)$$

The boundary condition $\theta(x \rightarrow \pm\infty) = 0$ implies that there is no reorientation induced in the sample for $|x| \rightarrow \infty$. In practice, we restrict the x domain of integration to a region $-x_{\max} \leq x \leq x_{\max}$, broad compared to the spot size ($x_{\max} \gg \omega_0$), and assume that $\theta = 0$ outside this domain. We proceed by discretizing the problem in x , thus transforming the partial differential equation (4.1) into a system of N ordinary differential equations for N functions $\theta_i(z)$, $i = 1, \dots, N$. The second derivative with respect to x is evaluated using a 3 points algorithm, coupling the

equations for θ_i to the equations for θ_{i+1} and θ_{i-1} . The assumption $\theta(x,z) = 0$ for $|x| > x_{\max}$ allows to truncate the infinite set of coupled equations for the $\theta_i(z)$, using $\theta_{i-1} = 0$ in the first equation and $\theta_{i+1} = 0$ in the last one. We then have to solve the system

$$d_{zz}^2 \theta_i + \frac{\theta_{i+1} - 2\theta_i + \theta_{i-1}}{(\Delta x)^2} + \beta_0 r \exp(-(x_i/\omega_0)^2) \frac{\sin\theta_i \cos\theta_i}{(1 - r \sin^2\theta_i)^{3/2}} = 0. \quad (4.3)$$

where $x_i = -x_{\max} + (i-1) \Delta x$, and $i = 1, \dots, N$.

Homeotropic boundary conditions are assumed at $z = 0$ and $z = d$, implying that $\theta_i(z=0) = \theta_i(z=d) = 0$ for all i . Δx is the step length introduced in the discretisation. We choose it small enough that the transverse profile of the solution does not depend on the value of Δx .

To integrate the equations, we have to start from a nonzero initial solution (since $\theta(x,z) = 0$ is a possible solution, the system always remains on this state if it is chosen as initial condition). Even then, integrating the system between $z = 0$ and $z = d$ and imposing that the reorientation angle is zero at the z -boundaries only leads to the zero solution. Instead, we seek solutions characterized by a symmetric longitudinal profile and impose that a maximum reorientation angle $\theta_{\max}$ should be reached in the middle of the sample. The corresponding intensity is deduced numerically by assuming a trial form of the transverse profile for $z = d/2$ and integrating the system of differential equations between $z = 0$ and $z = d/2$. The boundary conditions are not totally fixed: for $z = 0$, the angle is fixed to be zero, but its derivative is given as a parameter. At the other boundary $z = d/2$, the angle is given as a parameter and the derivative is fixed to zero (since the solution is symmetric around this point).

Integrating numerically the system of Eqs.(4.3) for various central peak intensities leads to various types of steady-state distributions for the reorientation angle $\theta(x,z)$. They can be characterized by 2 "mode indices" labeling the spatial structure in x and z . The fundamental mode, labeled as the $1_x 1_z$ solution, corresponds to the lowest intensity and is the only possible solution for β_0 close to the threshold intensity β_{th} (we use the plane wave threshold β_{th} obtained in Chapter III for reference).

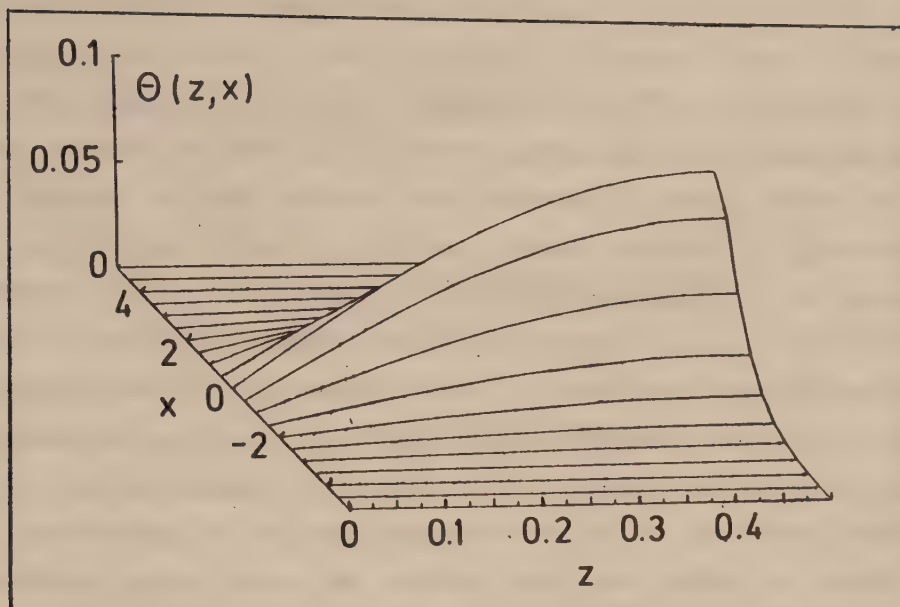


Fig. 4.1 Fundamental mode $1_x 1_z$ of the reorientation angle $\theta(z, x)$. z is in units of d , and only half of the sample is represented, since the solution is assumed to be symmetric about $z = 1/2$. x is in units of ω_0 . For this case, $d=250 \mu\text{m}$ and $\omega_0 = 200 \mu\text{m}$. $\theta_{\text{max}} = 10^{-1}$, the corresponding value of the intensity is $\beta_0 = 1.48$, in units of β_{th} .

Such a solution is illustrated in Fig. 4.1, for units scaled to the characteristic dimensions of the system (z and x are respectively scaled to d and ω_0). The maximum reorientation angle is reached in the middle of the sample, at $(x, z) = (0, 1/2)$. The longitudinal profile of θ is symmetric about $z = 1/2$ and it has no node between $z = 0$ and $z = 1$. A single maximum occurs at $z = 1/2$. For values of β_0 close to threshold, the z dependence of θ is well approximated by a $\sin(\pi z)$ function. This is to be expected from the approximation discussed in Chapter III since the reorientation angles are small. The transverse profile is symmetric about $x = 0$ and decreases monotonically to zero for increasing x . It is broader than the gaussian pump beam profile, due to the transverse correlations of the molecular reorientation through the sample.

When the input intensity is increased, the fundamental mode remains a solution, but saturation of the reorientation angle leads to a flatter longitudinal profile and a sharper increase of θ between its value 0 at the crystal's boundaries and its saturation value $\pi/2$ in a broad domain around $z = 1/2$. Higher order modes also become possible for high enough intensities, each mode m becoming possible when the intensity reaches its threshold value, typically given by the plane wave expression $m^2\beta_{th}$. For example, the 3_x1_z and the 1_x3_z modes shown in Fig. 4.2 are characterized by a fundamental distribution in one dimension, and a reorientation profile described by a third harmonic on the other. These modes have two nodes $\theta = 0$ between the boundaries, either in x or z direction, and are characterized by a maximum value of the reorientation angle much lower than θ_{max} corresponding to the same intensity in the fundamental mode. The peak intensities that allow to reach these higher order modes are far above threshold: this is to be expected since in the higher modes, the profiles have more structure and nodes (which corresponds to higher elastic energy) and the maxima in reorientation occur closer to the boundaries. If the intensity of the laser is further increased, even higher order modes can be reached.

The results of a systematic numerical study of Eq.(4.3) are summarized in graphical form in Fig. 4.3 and 4.4. Fig. 4.3 shows the dependence of the threshold peak intensity $\beta_{0,th}$ on the beam diameter ω_0 . $\beta_{0,th}$ decreases with increasing ω_0 and reaches the plane wave value β_{th} in the limit $\omega_0 \rightarrow \infty$. Increasing ω_0 while keeping the peak intensity constant is a way to increase the total power of the input beam. As pointed out in Chapter III, the peak threshold intensity is expected in this case to be lower, the broader the incident beam. The discussion of Chapter III, where the effect of the finite spot size on the threshold was calculated using a third-order nonlinearity and a square beam or a sech^2 profile is expected to still qualitatively hold when the full nonlinearity is kept and gaussian beams are considered. The dashed line of Fig. 4.3 reproduces the result of Chapter III expressed by (3.51). The discrepancy between the approximate analytical answer and the numerical results is due to the fact that the square profile is characterized by sharp edges and does not reproduce properly the wings of the gaussian profile. A better analytical expression for $\beta_{0,th}$ is found as the result of a linear calculation assuming a $\text{sech}^2(x/\omega_0)$ form of the laser beam, which leads to the expression (3.53) of the threshold intensity and corresponds to the dotted line of Fig. 4.3. This equation also shows that the

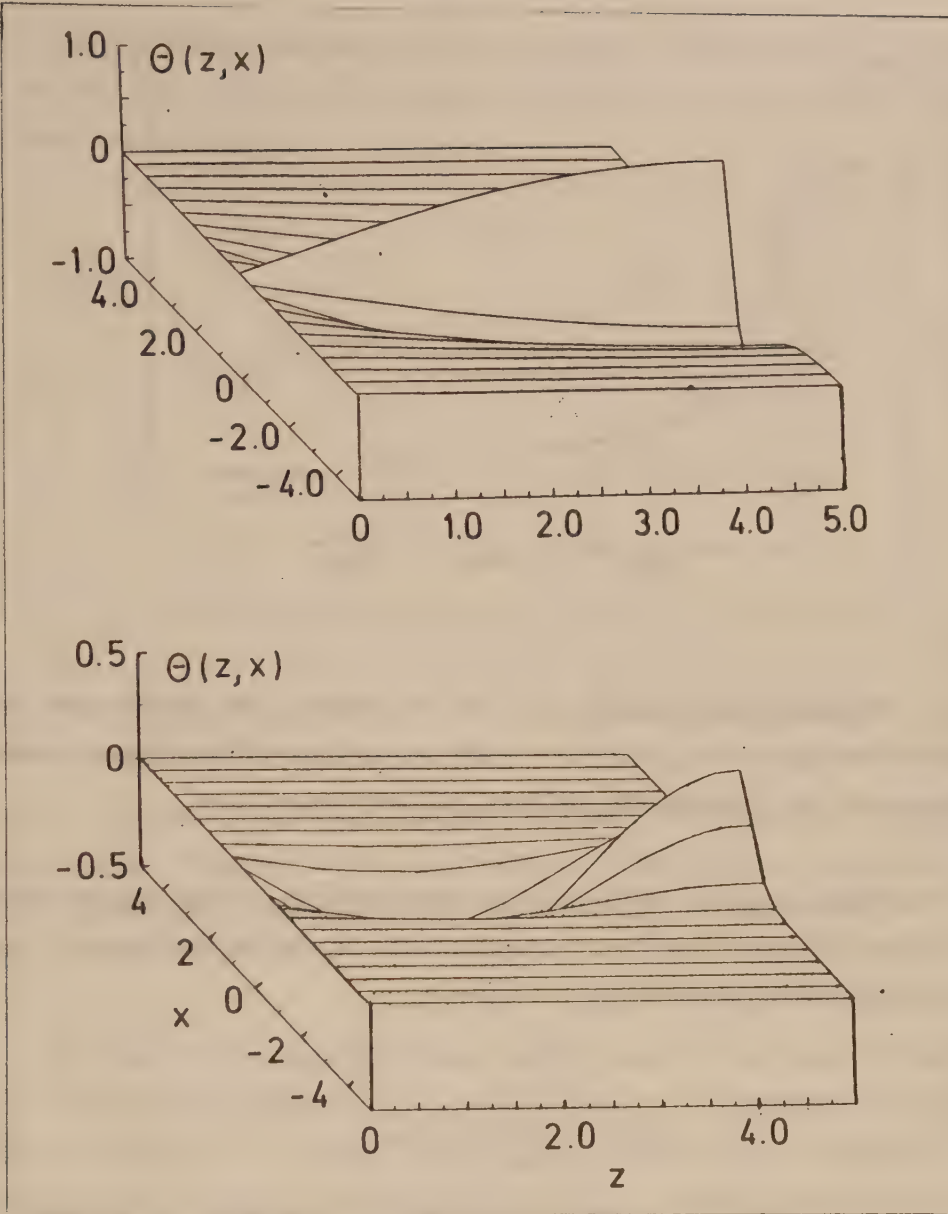


Fig. 4.2 Steady-state spatial distributions $\theta(z, x)$ for a sample length $d = 250 \mu\text{m}$ and a gaussian input beam waist $\omega_0 = 100 \mu\text{m}$. θ_{max} is fixed to 0.5 (a) $3_x 1_z$ mode corresponding to $\beta = 8.86$. (b) $1_x 3_z$ mode corresponding to $\beta = 11.87$.

integrated threshold intensity grows linearly with the beam diameter.

The maximum angle of reorientation θ_{max} is $\pi/2$, corresponding to the case where the directors are totally aligned with the field. θ_{max} is not easily measured

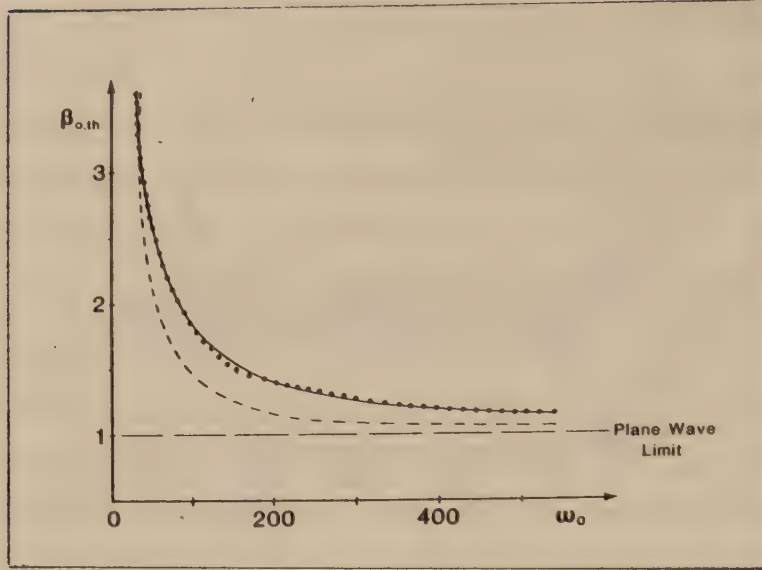


Fig. 4.3 Threshold peak intensity $\beta_{0,th}$ for the onset of the lowest mode, as a function of the spot size ω_0 [μm], for $d = 250 \mu\text{m}$ for three different beam profiles : gaussian (solid line), quadratic (dashed line) and sech^2 (dotted line).

in an experiment, contrary to the induced phase shift Φ_{nl} , which can be directly measured and is proportional to the integrated reorientation in the sample. In terms of the effective index of refraction, it is given by

$$\Phi_{nl} = \frac{2\pi}{\lambda} \int_0^d dz (n_{\text{eff}} - n_0) . \quad (4.4)$$

Once the reorientation angle distribution is known, it is easy to compute the effective refractive index and to perform the integration over z to obtain the nonlinear phase shift Φ_{nl} . The transverse profile of Φ_{nl} follows the profile of θ . Its maximum Φ_{max} is at $x = 0$, where the input beam has a maximum intensity. When the intensity is high, the whole sample is aligned with the field, and Φ_{max} reaches saturation. This is illustrated in Fig. 4.4(a), which gives $\Phi_{\text{max}}(\beta_0)$ in the case of a $1_x 1_z$ mode. Such large values of the nonlinear phase shift have actually been measured experimentally [Durbin et al. 1981a] and are a clear consequence of the giant nonlinearity of the NLC. The phase shift for the $1_x 1_z$ mode is represented as

function of the beam spot size for a given intensity in Fig. 4.4(b). $\Phi_{\max}$ is larger the broader the input beam, consistent with the fact that the OFT threshold decreases for increasing ω_0 .

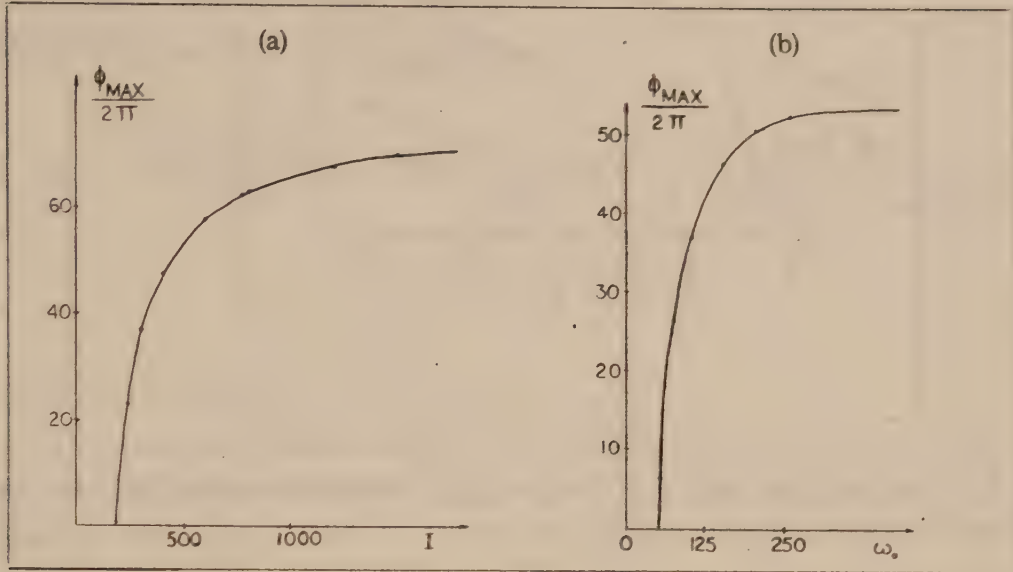


Fig. 4.4 Phase shift calculated for the $l_x l_z$ mode, for a sample of length $d = 250\mu\text{m}$ (a) as a function of the peak intensity I [W/cm²], $\omega_0 = 100\mu\text{m}$; (b) as a function of the beam diameter ω_0 [μm], for an intensity $I = 300$ W/cm².

The value of the phase shift being directly related to the integrated amplitude of the reorientation, a measure of $\Phi_{nl}(x)$ is a good way to distinguish between the different solutions: for a given intensity β_0 , the fundamental longitudinal mode corresponds to the highest reorientation angle. $\theta_{\max}$ decreases then with the order of the mode, and so does $\Phi_{\max}$. The profile $\Phi_{nl}(x)$ follows the transverse profile of the molecular reorientation, in other words it matches the transverse mode structure of the molecular reorientation. To illustrate the possibility of distinguishing between different modes by way of the nonlinear phase shift, we calculate Φ_{nl} for the first three modes of the molecular reorientation using the simplified one-dimensional model developed in the the preceding Chapter for the case of small dielectric anisotropy.

Fig. 4.5 displays these phase shifts as functions of the input intensity (given in units of the threshold intensity β_{th}). For intensities under saturation, the phase shift is effectively a decreasing function of the mode index and can be used to

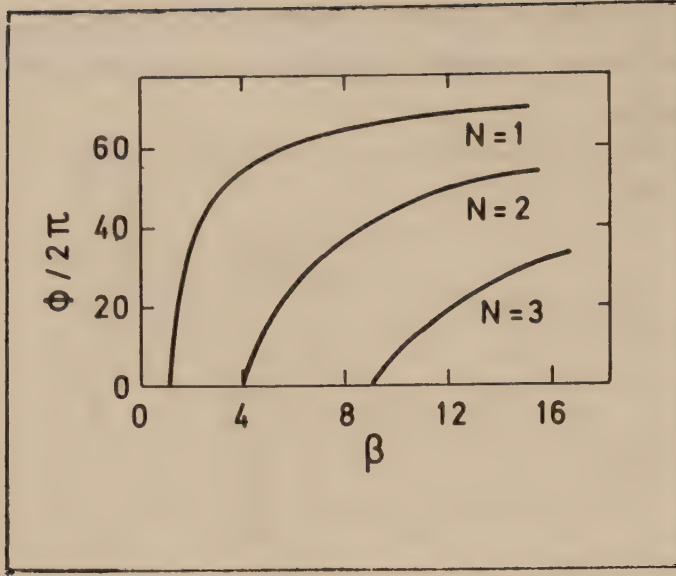


Fig. 4.5 Nonlinear phase shift as a function of the intensity (in units of β_{th}) for the first three longitudinal modes with $d = 250 \mu m$.

distinguish between different modes of the reorientation. When saturation dominates the system, the phase shift reaches a saturation value independent of the mode structure, since saturation is characterized by the whole medium being oriented parallel to the field. In this case, the phase shift does not allow to distinguish between different modes anymore.

IV.2 DYNAMICS

In the one-dimensional case studied in Chapter III, we found that all the longitudinal modes are unstable except for the fundamental one. To determine if this property remains true in the two-dimensional, fully nonlinear case, we now numerically analyze the general dynamic equation

$$\partial_{xx}^2 \theta + \partial_{zz}^2 \theta + \beta(x)r \frac{\sin\theta\cos\theta}{(1-r\sin^2\theta)^{3/2}} = \frac{\gamma}{\kappa} \partial_t \theta, \quad (4.5)$$

where $\beta(x)$ is a gaussian profile of the form (4.2).

We proceed by discretizing Eq.(4.5) in x and z , using as in the preceding Section a 3 points algorithm to evaluate the second-order derivatives. A grid of N points in x and M points in z is introduced, with integration step lengths Δx and Δz respectively. Eq.(4.5) is then reduced to a set of $N \times M$ coupled first-order differential equations in t for $\theta_{i,j}(t)$ of the type

$$\frac{\gamma}{\kappa} d_t \theta_{i,j} = \frac{\theta_{i+1,j} - 2\theta_{i,j} + \theta_{i-1,j}}{(\Delta z)^2} + \frac{\theta_{i,j-1} - 2\theta_{i,j} + \theta_{i,j+1}}{(\Delta x)^2} + \beta_0 r \exp(-(x_j/\omega_0)^2) \frac{\sin \theta_{i,j} \cos \theta_{i,j}}{(1 - r \sin^2 \theta_{i,j})^{3/2}}, \quad (4.6)$$

where $i = 1, \dots, M$ and $j = 1, \dots, N$.

The boundary conditions reflect the homeotropic alignment at $z=0$ and $z=d$, and the fact that no reorientation is induced for $|x| \rightarrow \infty$ (in practice for $|x| \gg \omega_0$). They are expressed by

$$\begin{aligned} \theta_{0,j} = \theta_{M+1,j} &= 0 \quad \text{for all } j \\ \theta_{i,0} = \theta_{i,N+1} &= 0 \quad \text{for all } i, \end{aligned} \quad (4.7)$$

We have performed a systematic study of (4.6) for various initial conditions. The main results of this analysis are now summarized:

- (1) For β larger than the threshold intensity, the higher modes are always unstable and evolve towards the fundamental $1_x 1_z$ solution.
- (2) If the input intensity is much larger than threshold, the system can be trapped in a higher mode for a very long time before evolving to the stable state. This is illustrated in Fig. 4.6, where the $3_x 1_z$ initial distribution is conserved for about 200 seconds. With even higher input intensities, we were able to trap the system in a higher mode for durations in excess of 1000 seconds.
- (3) If the peak intensity is abruptly changed when the system is in an unstable higher mode steady state, the system always switches to the corresponding fundamental mode, no matter how high the input intensity. In none of the cases studied were we able to force the system to switch to a higher mode. In Fig. 4.7 for example, the initial orientation is the $3_x 1_z$ solution, with $\beta_0 = 8.86 \beta_{th}$. When β_0

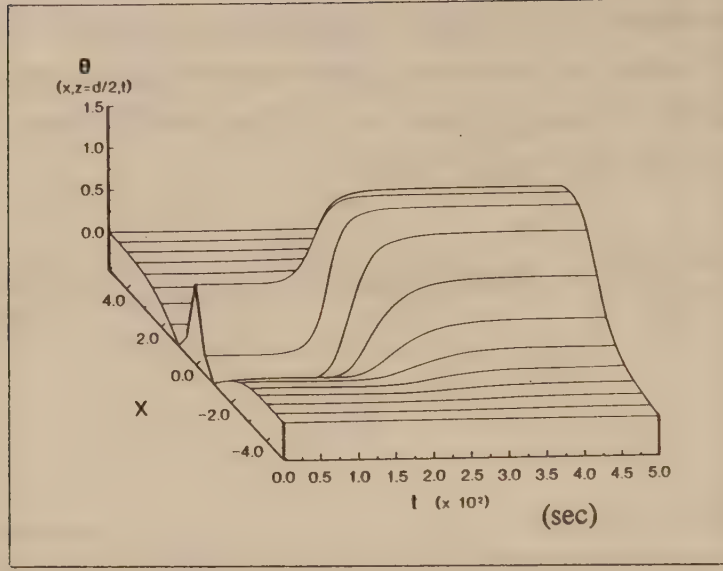


Fig. 4.6 Dynamical evolution of the $3_x 1_z$ mode for $\beta = 8.86 \beta_{th}$, $d = 250 \mu m$ and $\omega_0 = 100 \mu m$.

is suddenly increased to $12 \beta_{th}$, the corresponding $3_x 1_z$ solution is first reached, but soon the system escapes it to reach the fundamental mode.

(4) An alternative way of switching between different configurations is to induce a sudden change of the beam diameter, keeping the peak intensity fixed. This method is illustrated in Fig. 4.8. The initial situation is given by the fundamental mode corresponding to the intensity $\beta_0 = 1.92 \beta_{th}$. When ω_0 is reduced by a factor 2, θ decays to zero, since β_0 is now under the threshold corresponding to this beam waist. On the contrary, when ω_0 is doubled, θ evolves to a broader fundamental distribution, since the corresponding peak threshold is lowered. As already discussed in Chapter III, modifying ω_0 corresponds to a change in the integrated input intensity of the laser beam, driving the system into a different orientational distribution.

We have studied the case where the field is switched on at time $t = 0$, by integrating the dynamical equation with a uniform very small initial condition $\theta(x,z,t=0) = \epsilon$, where $\epsilon \ll 1$. The time constant of the system is inversely proportional to $(\beta - \beta_{th})$ as was the case in the linear calculation of the preceding

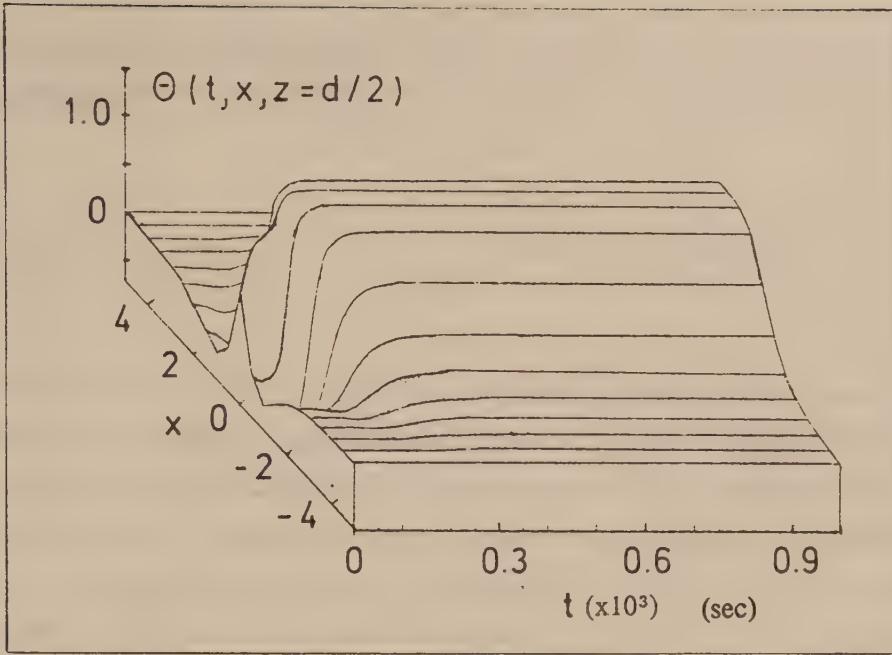


Fig. 4.7 Effect of a sudden increase of the input intensity, for the $3_x 1_z$ mode. β is increased from 8.86 to 12. The values of the parameters are $d = 250 \mu\text{m}$ and $\omega_0 = 100 \mu\text{m}$.

Chapter (Eq.(3.20)). Again, this is a signature of critical slowing-down occurring near a transition point, characteristic of a second-order phase transition.

We summarize the results of this Chapter by noting that the mode structure found analytically is still present in a numerical simulation, and that the transverse correlations of the medium induce non-monotonic transverse profiles in the molecular reorientation. The number of available solutions increases with the incident intensity, but only the fundamental distribution is stable for very long times. We never succeed in switching the system from $\theta(x, z) \cong 0$ to a higher mode, even with very high laser intensities. This conclusion has to be revised in the presence of noise: we see in the next Chapter that higher modes of the molecular reorientation can be obtained in a Freedericksz transition, when thermal fluctuations are introduced in the system.

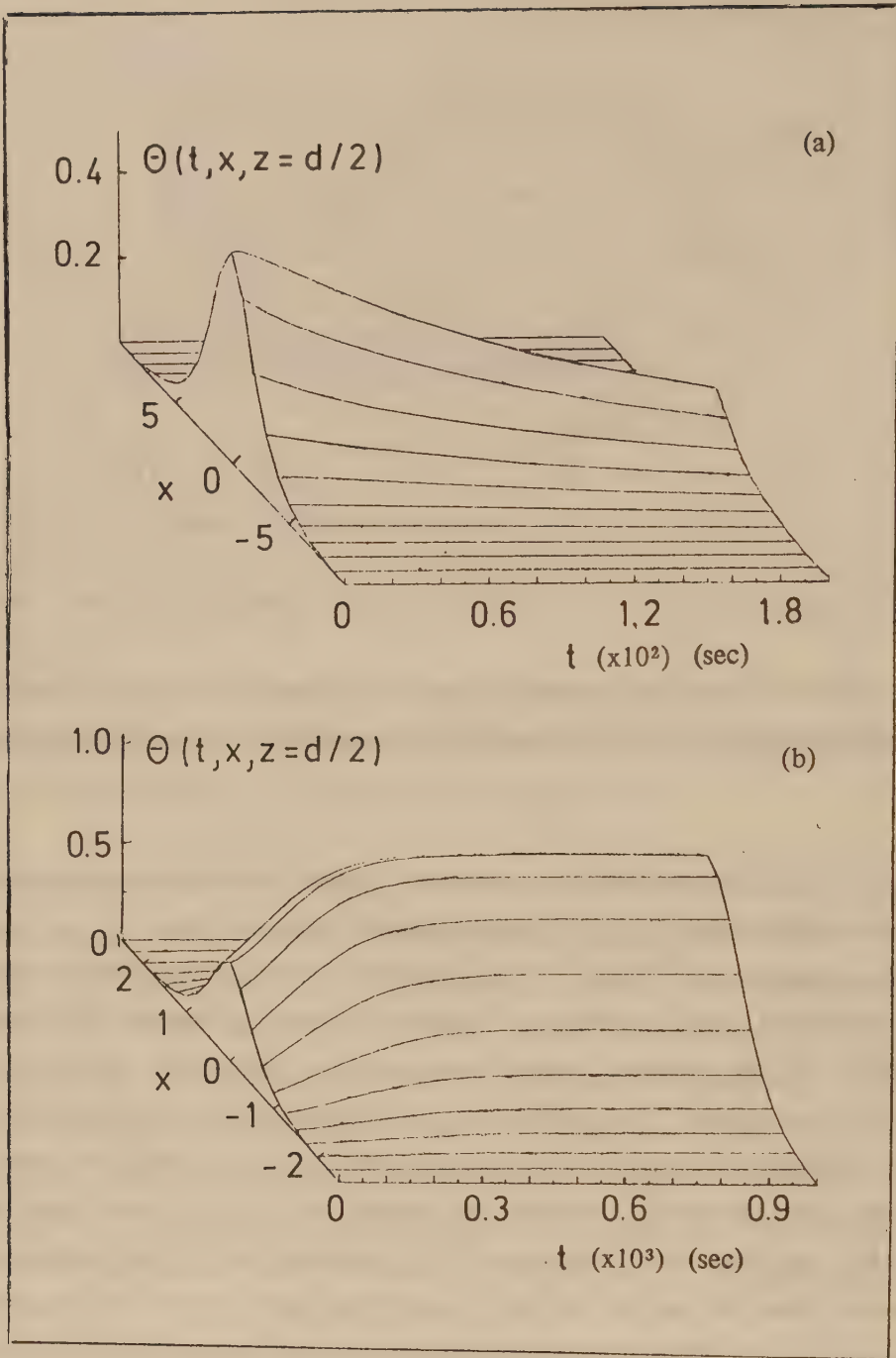


Fig. 4.8 Effect of a variation of the input beam spot size on the molecular reorientation. The initial situation corresponds to $\omega_0 = 100 \mu\text{m}$, $\beta = 1.92 \beta_{\text{th}}$ (a) ω_0 is suddenly reduced to $50 \mu\text{m}$; (b) ω_0 is suddenly increased to $200 \mu\text{m}$.

V. STOCHASTIC DESCRIPTION OF THE OPTICAL FREEDERICKSZ TRANSITION

V.1 INTRODUCTION

So far, we have treated the dynamics of the optical Freedericksz transition in a completely deterministic way, and found that for intensities under threshold, the only possible molecular orientation is characterized by a uniform angle $\theta(x,z,t) = 0$. For intensities above threshold, several different inhomogeneous θ distributions are possible. In analogy to the potential V introduced in Chapter III for the case of small reorientation, all these solutions can be discussed in terms of a generalized potential $U(\theta)$. Each possible director angle distribution $\theta(x,z,t)$ corresponds to an extremum of this potential. $U(\theta)$ is not easily visualized, since being a function of the field $\theta(x,z,t)$, it has infinite dimension, but it is still a useful concept in discussing the transition from one state to another. For intensities below threshold, the only minimum of $U(\theta)$ is for $\theta(x,z,t) = 0$. It becomes a local maximum when the intensity is increased past threshold. The fundamental inhomogeneous mode corresponds then to a minimum of $U(\theta)$. As the intensity is further increased, new minima appear each time a higher mode becomes possible. When the input intensity is large enough to allow higher order solutions beside the fundamental nonhomogeneous distribution, the deterministic analysis of Chapters III and IV predicts that the system is invariably driven into the potential corresponding to the fundamental mode. To determine if this remains true in a real system in the presence of thermal fluctuations, or whether it is only a feature of the deterministic model, we now introduce internal fluctuations in the medium and describe the transition as a stochastic process.

This Chapter discusses a stochastic model of the Freedericksz transition, including internal fluctuations of the molecular alignment due to thermal agitation. We restrict our analysis to the simple model of a one-dimensional system and study specifically the transient dynamics of the director orientation when the laser is switched on. A similar case is discussed by Sagues and San Miguel [Sagues and San Miguel 1985], who study a stochastic model of the one-dimensional Freedericksz transition in a static magnetic field. They describe the internal fluctuations of the

liquid crystal by an additive noise term, while the random fluctuations of the magnetic field lead to a multiplicative noise component. They perform a third-order calculation in the reorientation angle θ , thus limiting their model to low intensities of the magnetic field. Their approximations lead to a potential U which is a function of the maximum θ_m of the reorientation angle. Because of their multiplicative character, the fluctuations of the magnetic field shift the position of the minima of the potential, and hence the position of the threshold of the Freedericksz transition, the critical value of the field being reduced by a quantity proportional to the strength of correlations of the fluctuations of the magnetic field.

In contrast to this work, we consider in our model internal fluctuations only, but keep in our model the full nonlinearity to all orders in θ . This allows the study of the dynamics of higher order modes. We discuss the case of a plane wave incident laser field illuminating at normal incidence a homeotropically aligned sample of NLC of longitudinal dimension d and infinite transverse dimensions. The input laser beam is polarized along the x direction; we assume that only fluctuations in the x - z plane are amplified and that the system is stable against fluctuations in the x - y plane. The problem is then homogenous in the transverse dimensions and reduced to a one-dimensional situation, where only the spatial longitudinal coordinate z is relevant. Introducing internal fluctuations, the dynamics of the molecular reorientation angle $\theta(z,t)$ is governed by the Langevin equation,

$$\partial_t \theta = \frac{\kappa}{\gamma} \left[\partial_{zz}^2 \theta + \beta r \frac{\sin \theta \cos \theta}{(1 - r \sin^2 \theta)^{3/2}} \right] + \eta(z,t) , \quad (5.1)$$

where a stochastic force η has been added to the deterministic equation (2.12) derived in Chapter II. The x dependence of the noise is neglected, since we assume a plane wave model and neglect the possible transverse inhomogeneities in the reorientation angle. We assume the average value of the thermal fluctuations to be zero, and the thermal fluctuations at the point (z,t) to be uncorrelated to the fluctuations acting at (z',t') . Specifically, η is then taken as a gaussian white noise of zero mean and delta correlated

$$\langle \eta(z,t) \eta(z',t') \rangle = 2\epsilon \delta(t-t') \delta(z-z') , \quad (5.2)$$

where ϵ is a constant which measures the strength of the fluctuations.

The deterministic part on the right-hand side of the Langevin equation corresponds to the variation of the free energy density in the sample as the director angle θ is varied. This is easily seen by expressing the free energy F for this specific case as

$$F = A \int dz \mathcal{F} = A \int dz \frac{1}{2} \kappa [(\partial_z \theta)^2 - 2\beta(1-r\sin^2\theta)^{-1/2}], \quad (5.3)$$

where A is the transverse area of the liquid crystal sample. Defining the quantity $\frac{\delta \mathcal{F}}{\delta \theta}$ in a variational sense

$$\frac{\delta \mathcal{F}}{\delta \theta} = \frac{\partial \mathcal{F}}{\partial \theta} - \frac{d}{dz} \frac{\partial \mathcal{F}}{\partial (\partial_z \theta)}, \quad (5.4)$$

the Langevin equation (5.1) can be written as

$$\frac{\partial \theta}{\partial t} = -\frac{1}{A\gamma} \frac{\delta \mathcal{F}}{\delta \theta} + \eta(z,t), \quad (5.5)$$

as advertised. In the second Section of this Chapter, we solve numerically a discretized form of equation (5.1) for a large number of realizations of the noise term η , studying the transition from the uniform homeotropic orientation to a nonuniform θ distribution. This allows to discuss the effects of thermal fluctuations on the behaviour of the system in a statistical sense. In particular, the characteristic average time for the onset of a nonhomogenous molecular distribution is determined by averaging the result of computer simulations of the stochastic process over a large number of realizations.

Another equivalent way to proceed is to use the Fokker-Planck equation associated to the Langevin equation (5.1). This equation governs the transition probability to a specified state characterized by a given $\theta(z)$ distribution from an initial state $\theta_0(z)$. The Fokker-Planck equation is a dynamical equation for this probability $W(\theta)$, given an initial condition θ_0 . It involves a drift term corresponding to the deterministic part of the process and a diffusion term related to the stochastic force η . The Fokker-Planck equation corresponding to the Langevin equation (5.1) is

$$\partial_t W(\theta) = -\partial_\theta [h(z,t)W(\theta)] + \partial_{\theta\theta}^2 [g(z,t)W(\theta)] , \quad (5.6)$$

where $W(\theta)$ is the probability to induce a Freedericksz transition to a molecular orientational state described by θ . The drift and diffusion coefficients h and g can be deduced from the Langevin equation by averaging the stochastic variables θ and θ^2 as

$$h(z,t) = \lim_{\tau \rightarrow 0} \frac{1}{\tau} \langle \theta(t+\tau) - \theta(t) \rangle$$

$$g(z,t) = \frac{1}{2} \lim_{\tau \rightarrow 0} \frac{1}{\tau} \langle [\theta(t+\tau) - \theta(t)]^2 \rangle . \quad (5.7)$$

Performing these limits explicitly (the details are given in Section 3 for the discrete case), the explicit forms of the coefficients h and g are found to be

$$h(z,t) = \frac{\kappa}{\gamma} \left[\partial_{zz}^2 \theta + \beta r \frac{\sin\theta \cos\theta}{(1-r\sin^2\theta)^{3/2}} \right]$$

and

$$g(z,t) = \epsilon . \quad (5.8)$$

Section 3 discusses the stationary solution $W_{st}(\theta)$ of the Fokker-Planck equation. For our model, it is given by

$$W_{st}(\theta) = \mathcal{N} \exp \left[-\frac{F(\theta)}{kT} \right] , \quad (5.9)$$

where $F(\theta)$ is the free energy of the liquid crystal for the molecular orientation $\theta(z)$, k is the Boltzmann constant, T the temperature in °K and $\mathcal{N}$ a normalization constant. When the steady-state is reached, the most probable molecular configuration corresponds then to a minimum of the free energy. Expression (5.9) shows that the distribution $\theta(z)$ corresponding to the lowest free energy is the most likely to occur in a transition.

V.2 LANGEVIN APPROACH - NUMERICAL SOLUTION

To numerically solve the Langevin equation (5.1), it must be put into a discretized form approximating the solution for the field $\theta(z,t)$ by a set of M values $\{\theta_j(t)\}$, at the points $\{z_j\}$. Considering first the deterministic part of Eq.(5.1), we discretize it by dividing the length of the crystal in $M+1$ elements of length $\Delta z = \frac{d}{M+1}$. This procedure leads to a system of M coupled deterministic equations for $\theta_i(t)$, $i=1,\dots,M$.

$$\frac{\gamma}{\kappa} \partial_t \theta_i = \frac{\theta_{i+1} - 2\theta_i + \theta_{i-1}}{(\Delta z)^2} + \beta r \frac{\sin \theta_i \cos \theta_i}{(1 - r \sin^2 \theta_i)^{3/2}} \equiv f_i^{\text{det}} . \quad (5.10)$$

The homeotropic boundary conditions are satisfied by setting $\theta_{i-1} = 0$ in the first and $\theta_{i+1} = 0$ in the last equation. A standard one-step integrator is used to perform the time integration, the integration interval $(t-t_0)$ being discretized by introducing N values $t_j = j \Delta t$, $j = 1,\dots,N$. The director angle $\theta_i(t_{n+1})$ can thus be expressed in function of $\{\theta_j(t_n)\}$ as

$$\theta_i(t_{n+1}) = \theta_i(t_n) + \frac{\kappa}{\gamma} \Delta t f_i^{\text{det}}(t_n) . \quad (5.11)$$

The initial condition $\theta_i(t=t_0) = \theta_{i,0}$ completely determines the solution. The step lengths Δt and Δz must be chosen small enough to lead to solutions that are independent of their fixed values.

After introducing the stochastic force $\eta(z,t)$ into the Langevin equation, Eqs.(5.10) become

$$\partial_t \theta_i = \frac{\kappa}{\gamma} f_i^{\text{det}}(t) + \xi_i(t) , \quad (5.12)$$

where $\xi_i(t)$ is a gaussian noise force acting on the variable θ_i only. Consistently with

(5.2) the various $\xi_i(t)$ have zero mean and are delta correlated,

$$\langle \xi_i(t) \xi_j(t') \rangle = 2D \delta_{i,j} \delta(t-t'), \quad (5.13)$$

where $D = \epsilon/d$ is independent of i and j .

To be able to write the solution $\theta_i(t_{n+1})$ as a function of the $\{\theta_j(t_n)\}$, the noise force must be discretized, too. We proceed by introducing a new stochastic variable

$$\xi'_i(t_n) = \frac{1}{\sqrt{D}} \xi_i(t = t_n), \text{ defined for discrete times } t_n \text{ and scaled to } \sqrt{D}. \text{ Using the}$$

discretized form of the first derivative, the Langevin equation (5.12) can then be rewritten as a function of the ξ'_i as

$$\frac{[\theta_i(t_{n+1}) - \theta_i(t_n)]}{\Delta t} = \frac{\kappa}{\gamma} f_i^{\text{det}}(t_n) + \sqrt{D} \xi'_i(t_n). \quad (5.14)$$

The correlations of the variables ξ'_i are expressed by the discretized form of (5.13), leading to $\langle \xi'_i(t_n) \xi'_j(t_m) \rangle = 2/\Delta t \delta_{i,j} \delta_{n,m}$. The last step of the discretization rescales the noise term to $\frac{1}{\sqrt{\Delta t}}$, introducing the new stochastic quantity μ_i

$$\mu_i(t_n) = \sqrt{\Delta t} \xi'_i(t_n). \quad (5.15)$$

From the characteristics of ξ'_i , it follows that μ is a stochastic variable of zero mean, and of correlations satisfying $\langle \mu_i(t_n) \mu_j(t_m) \rangle = 2 \delta_{i,j} \delta_{n,m}$. The solution of the Langevin equation (5.12) is then given by a one-step integrator as

$$\theta_i(t_{n+1}) = \theta_i(t_n) + \frac{\kappa}{\gamma} \Delta t f_i^{\text{det}}(t_n) + \sqrt{D \Delta t} \mu_i(t_n), \quad (5.16)$$

calculated from a specified set of $\theta_i(t=t_0)$.

The delta-correlated noise is an idealized representation of a physical force whose correlation time is short compared to all characteristic times of the system. To simulate the physical noise μ_i on the computer, we introduce a characteristic time t_c

which is much smaller than any characteristic times of the deterministic system, but larger than the time step Δt introduced in the discretization. μ_i is not markovian in this approximation. Rather, it is modeled by weakly colored noise of mean and correlations given by

$$\langle \mu_i(t_n) \rangle = 0$$

$$\langle \mu_i(t_n) \mu_j(t_m) \rangle = 2 \exp \left[- \frac{|(t_n - t_m)|}{t_c} \right]. \quad (5.17)$$

With this form, all correlations between different times are washed out for time differences of the order or larger than the smallest characteristic time of the deterministic system, as they should. But on the time scale of Δt , noise correlations are still present, allowing a better numerical convergence of the one-step predictor integration.

Using gaussian random variables R centered on zero and characterized by correlations $\langle R^2 \rangle = 2$, the noise $\mu_i(t_n)$ is constructed as

$$\mu_i(t_1) = R(t_1)$$

$$\mu_i(t_n) = \exp(-\Delta t/t_c) \mu_i(t_{n-1}) + \sqrt{1 - \exp(-2\Delta t/t_c)} R(t_n), \quad n = 2, \dots, N \quad (5.18)$$

This form of the noise satisfies the mean and correlations (5.17), as can easily be seen by direct calculation. To construct the gaussian variables $R(t_n)$, we use a random number generator to produce L random numbers r_ℓ between 0 and 1, and obtain R as

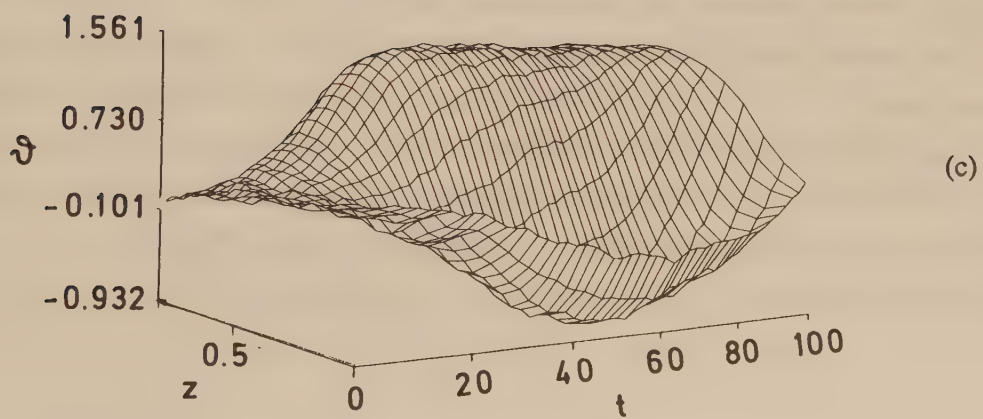
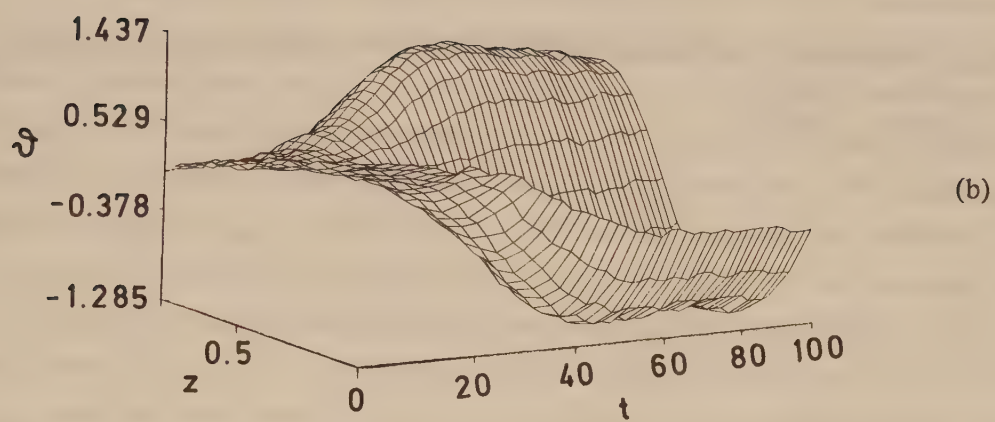
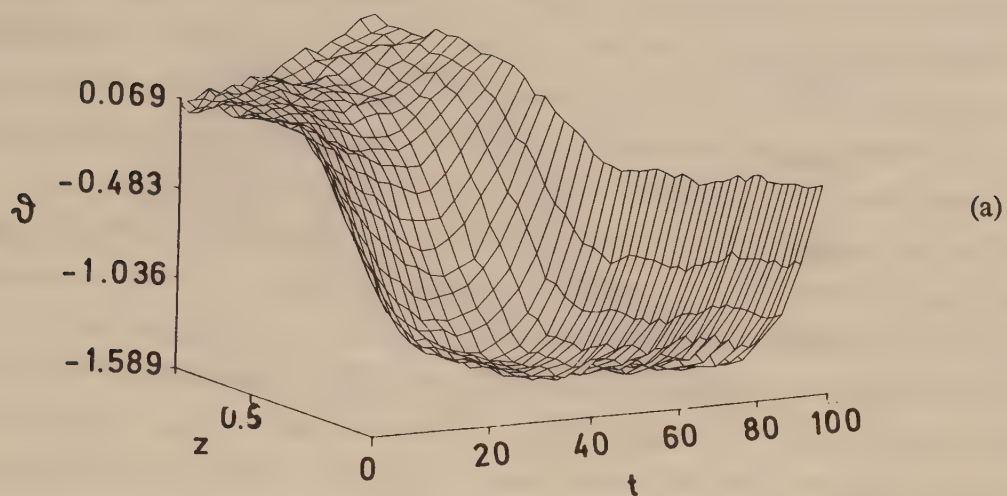
$$R = \sqrt{24/L} \sum_{\ell=1}^L (r_\ell - 1/2). \quad (5.19)$$

For each i and n , R is constructed using L new random numbers. The central limit theorem guarantees that the random variables R are gaussian distributed for large L . In practice, it is sufficient to take $L = 10$.

In a series of numerical experiments, we have switched the laser at time $t = 0$ from zero to a fixed intensity larger than threshold. The initial molecular orientation is homogenous and characterized by $\theta(z,t=0) = 0$. The time evolution of θ is followed during 100 seconds. Following the laser switch on, the Freedericksz transition occurs, with the internal fluctuations amplified to drive the system away from the unstable homogenous initial state into a nonhomogenous distribution. We have computed the solution (5.16) of the Langevin equation for a large number of realizations of the stochastic process, keeping the deterministic parameters fixed. Averaging over all realizations allows to determine the statistics of the switch-on time τ for the onset of a nonhomogenous reorientation distribution.

The first series of realizations of the stochastic process was performed for a constant laser intensity β equal to 10 times the plane-wave threshold of the OFT. The most remarkable result of this numerical experiment is that in a few cases, the second spatial mode is reached, in contrast to what happens in a deterministic description (Chapters III and IV). This second mode is very close to the second mode of the elliptic sine function, which is the approximate solution obtained in Chapter III for the case of small dielectric anisotropy. (In the numerical calculations performed here, the dielectric anisotropy r was fixed to $r = .23$, corresponding to a sample of MBBA). In some cases, the system was found to evolve towards the second longitudinal mode and to sustain this reorientation distribution for times at least as large as 100 seconds. A few examples of the possible dynamical behaviours of $\theta(z,t)$ are illustrated in Fig. 5.1 for $D = 10^{-4}$, Δt and t_c being respectively fixed to the values $1/15$ and 10^{-2} seconds. Case (a) corresponds to the most common dynamical evolution of θ : the unstable zero solution evolves towards the fundamental inhomogenous longitudinal mode, where saturation occurs in the reorientation (a large part of the longitudinal profile of θ reaches the maximum value $\pm\pi/2$). The sign of θ is of course physically irrelevant, i.e. $\theta(z,t)$ is physically equivalent to and undistinguishable from $-\theta(z,t)$. Positive and negative angles of reorientation have been observed to appear with equal frequencies within the statistical uncertainty of our numerical experiment.

Case (b) represents the situation already mentioned where the fluctuations drive the system into the second spatial mode. The longitudinal profile at time $t = 100$ sec is not always perfectly symmetric as would be expected from a deterministic model, suggesting that for those specific realizations, the second mode is only a transient



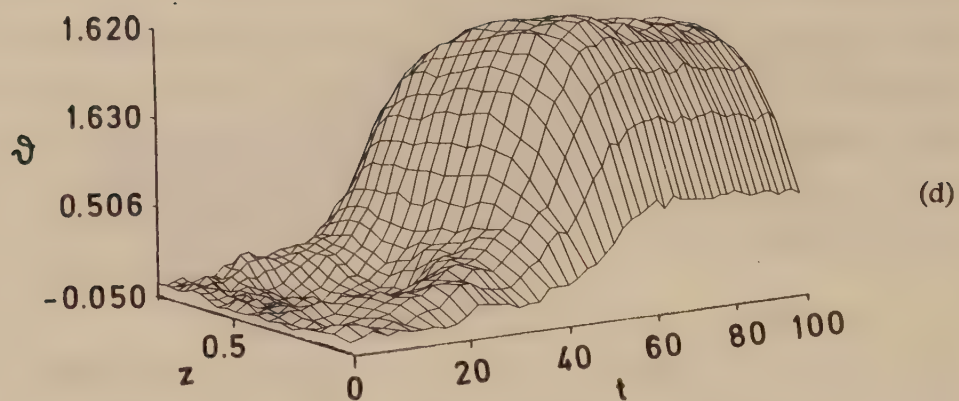


Fig.5.1 Dynamical behaviours of $\theta(z,t)$ for $D = 10^{-4}$, $d = 250 \mu\text{m}$ and $\beta = 10 \beta_{\text{th}}$. $\Delta t = \frac{1}{15}$ and $t_c = 10^{-2}$.

solution and that for longer integration times, the fluctuations will drive the system back into the fundamental mode. Such a situation is illustrated in Fig. 5.1(c), where the second mode is clearly excited in a transient regime before the fundamental distribution is reached. Still, it is interesting to see that contrary to what might be expected in view of the deterministic analysis, the OFT can in principle lead to higher modes of the molecular orientational distribution than the fundamental one.

With an input intensity ten times above threshold, the third mode of reorientation can in principle be reached. In the 200 realizations studied in this experiment, the third mode never appears clearly, but in a few cases, the very first part of the evolution showed a tendency to evolve towards such a mode. In Fig. 5.1(d), we see that for small times, the longitudinal profile is characterized by two minima, one on each side of $z = d/2$, typical of a third-order mode reorientation. This profile is however rapidly washed out and the clear evolution towards the fundamental mode takes place.

To determine the switch-on time τ characterizing the onset of a nonhomogenous reorientation, we have compared the longitudinal distribution of $\theta(z,t)$ with the different possible modes of the elliptic sine derived in (3.6) from a deterministic calculation in the small dielectric anisotropy limit: For a fixed input intensity β , the derivation of Chapter III and in particular Eq.(3.8) shows that the maximum reorientation angle $\theta_{\max,m}$ is different for each mode, providing that saturation is not reached. Furthermore, the position z_m of the maximum in each sn-mode is easily determined from the general properties of the elliptic sine function. We can then compare the values of the numerical solution $\theta(z_m, t_n)$ at various times t_n with $\theta_{\max,m}$, and use as a criterion for the onset of mode m the condition that $\theta(z_m, t_n)$ reaches the value $1/2 \theta_{\max,m}$ at z_m . The switch-on time τ is then defined as the corresponding t_n . This procedure is illustrated in Fig. 5.2

Averaging the values of τ over all realizations of the stochastic process allows to determine the statistics of the switch-on times to the different modes. For the present case where β is fixed to $10 \beta_{th}$, the statistics for the onset of higher order spatial modes were performed using 200 realizations of the stochastic process. The results are summarized in Fig. 5.3. Histogram (a) is constructed with 194 events that lead to the fundamental mode, out of the 200 realizations computed for $D = 10^{-4}$. The mean value for τ_1 corresponds to $\langle \tau_1 \rangle = 34.45$ sec, with a dispersion of $\Delta = 8.19$

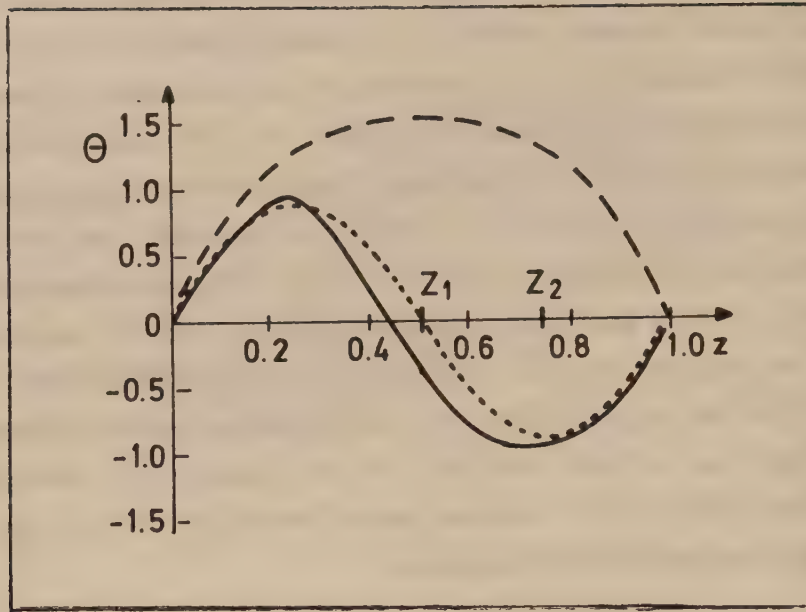


Fig. 5.2 Criterion used to determine the onset of mode m , for $m = 1$ and 2 . The solid line corresponds to the numerical solution $\theta(z)$ for a fixed time t . The dashed (dotted) line displays the fundamental (second) mode of the elliptic sine. Comparing the three curves at $z = z_1$ and $z = z_2$ allows to distinguish between the first and the second mode.

sec. $\left[\text{The dispersion } \Delta \text{ is defined as } \sqrt{1/N \langle (t - \langle t \rangle)^2 \rangle} \right]$

To analyze the effect of the noise strength on τ_1 , a second series of 200 realizations was integrated for the smaller value $D = 10^{-5}$. In this case, the switch-on time τ_1 characteristic of the onset of the fundamental mode is larger. For smaller noise, the system needs on the average a longer time to leave the unstable homogenous state $\theta(z) = 0$ and to build the fundamental nonhomogenous distribution. (This has already been mentioned in the deterministic case, when the dynamics of the one-dimensional system was studied in the small dielectric anisotropy limit.) The results of the 194 realizations leading to the fundamental mode are reported in Fig. 5.3(b). The average value of τ_1 is now 44.61 sec, and the dispersion is $\Delta = 8.56$ sec. Reducing D by a further factor of 10 from its previous value to $D = 10^{-6}$ leads

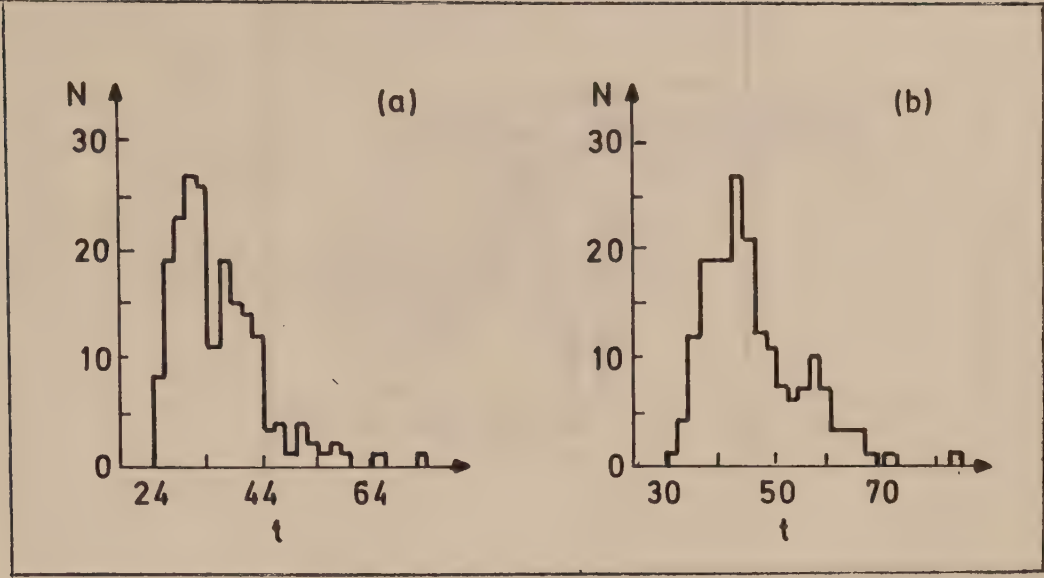


Fig. 5.3 Distribution of the switch-on time τ_1 to the first nonhomogenous mode of the reorientation. Each histogram is constructed from a series of 200 realizations of the stochastic noise. t is given in seconds. (a) $D = 10^{-4}$. (b) $D = 10^{-5}$. Note the different time scales on these two Figures.

to a still increased value of the switch-on time τ_1 . In this case, $\langle \tau_1 \rangle = 51.73$ sec, with a dispersion $\Delta = 8.72$ sec.

The same numerical experiment was repeated for a fixed value of $D = 10^{-4}$ and a higher laser intensity $\beta = 20 \beta_{th}$. In this case, higher order modes are expected to be reached with a higher frequency. We found indeed that out of 200 realizations of the stochastic process, 160 corresponded to a Freedericksz transition to the fundamental inhomogenous θ distribution, the 40 remaining cases leading to the second mode distribution. The switch-on time calculated in this series was $\langle \tau_1 \rangle = 16.87$ sec (cf. Fig. 5.4). The dispersion of the values around this mean value is noticeably smaller, $\Delta = 3.09$ sec.

Comparing this series of realizations to the results $\beta = 10 \beta_{th}$ shows that for a given noise strength, it is much more probable to see a Freedericksz transition to a higher mode when the intensity of the laser is large. Furthermore, the first part of the evolution is characterized by a richer structure in this last case. The number of

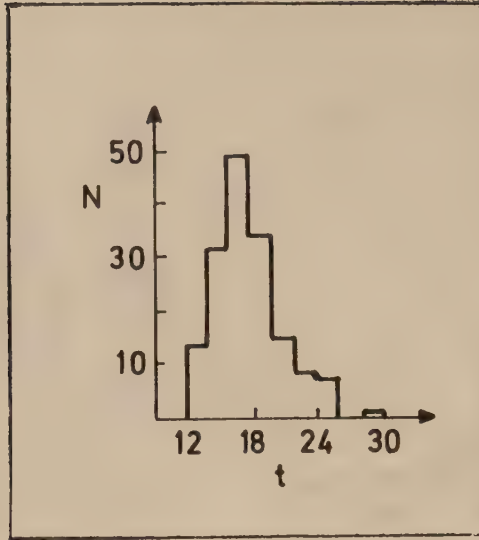


Fig. 5.4 Distribution of the switch-on time τ_1 for $\beta = 20 \beta_{th}$.

realizations for which an evolution towards a higher mode (third or even fourth) shortly appears in a transient regime is increased with the input intensity. For example, the realization reported in Fig 5.5 clearly displays a transient evolution from the unstable zero state to the third mode, with a final evolution towards the fundamental distribution. This suggests that the higher modes of the molecular reorientation can be observed with high enough intensities. To determine the asymptotic steady state of the molecular reorientation, a probabilistic description of the optical Freedericksz transition is discussed in the following Section.

V.3 FOKKER-PLANCK APPROACH

The preceding Section showed that under the influence of thermal fluctuations, a Freedericksz transition can occur not only to the fundamental nonhomogenous state, but also to several higher modes of the molecular reorientation distribution. We now present a probabilistic description which allows to discuss directly the statistics of all possible orientational states $\theta(z,t)$ without having to perform a high number of independent realizations of the stochastic process. The probability to observe a transition to an arbitrary state of the molecular reorientation $\theta(z,t)$ is governed by a

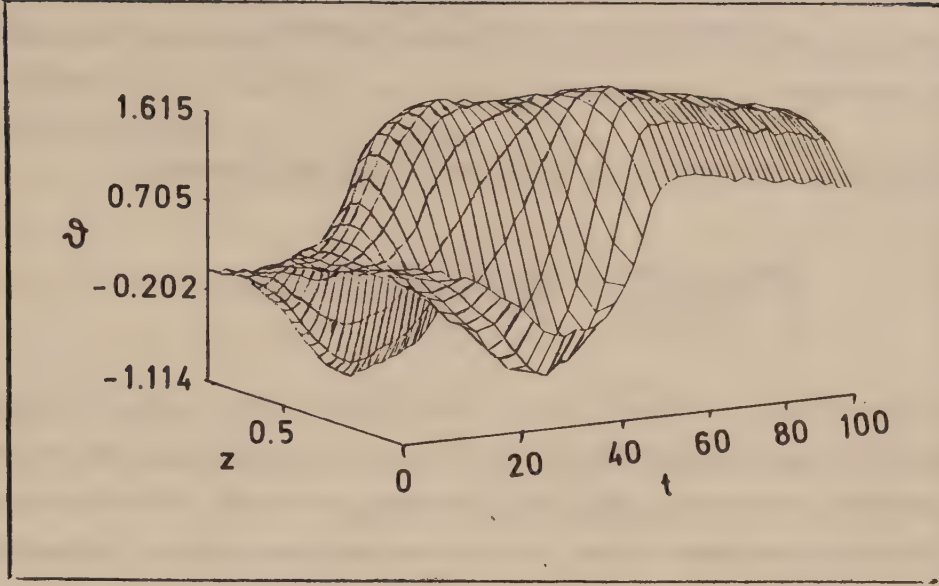


Fig. 5.5 Dynamics of the Fredericksz transition for $\beta = 20 \beta_{th}$. The noise strength is $D = 10^{-4}$.

Fokker-Planck equation of the type introduced in Eq.(5.6). Since we describe the problem in a discretized form of the spatial variable z , a particular state corresponding to the field θ is represented by the set of M functions $\{\theta_j(t)\}$. The probability $W(\{\theta_j\})$ to find a state $\{\theta_j\}$ obeys a Fokker-Planck equation involving M different drift coefficients h_i and M^2 diffusion coefficients $g_{i,j}$. As mentioned in Section V.1, these can be calculated from the Langevin equation, averaging the stochastic variable θ_i and $\theta_i\theta_j$.

$$h_i = \lim_{\tau \rightarrow 0} \frac{1}{\tau} \langle \theta_i(t+\tau) - \theta_i(t) \rangle$$

$$g_{i,j} = \frac{1}{2} \lim_{\tau \rightarrow 0} \frac{1}{\tau} \langle (\theta_i(t+\tau) - \theta_i(t))(\theta_j(t+\tau) - \theta_j(t)) \rangle. \quad (5.20)$$

To perform the calculations of these coefficients, we first express the difference $\theta_i(t+\tau) - \theta_i(t)$ as an integral, using the integral form of the Langevin equation

$$\theta_i(t+\tau) - \theta_i(t) = \int_t^{t+\tau} dt' \left[\frac{\kappa}{\gamma} f_i^{\text{det}}(t') + \eta_i(t') \right]. \quad (5.21)$$

The deterministic force depends on θ_i and can be expanded according to

$$f_i^{\text{det}}(t') = f_i^{\text{det}}(t) + \frac{\partial}{\partial \theta_i} f_i^{\text{det}}(t) [\theta_i(t') - \theta_i(t)] + \dots \quad (5.22)$$

The same procedure is followed for the quantity $[\theta_i(t') - \theta_i(t)]$, leading to multiple integrals. The noise describing the thermal fluctuations is a white noise of zero mean and correlations expressed by $\langle \eta_i(t) \eta_j(t') \rangle = 2\epsilon / \Delta z \delta_{i,j} \delta(t-t')$, the discretized form of (5.2). Taking the statistic averages, only terms involving products of the form $\eta_i(t') \eta_i(t'')$ contribute to the integral, all others being zero due to the properties of the white noise η_i . The calculation can easily be performed, but it leads to rather long expressions. We give only the final result, since the method is standard [Risken 1984].

$$h_i = \frac{\kappa}{\gamma} f_i^{\text{det}}(t)$$

$$g_{i,j} = \frac{1}{\Delta z} \epsilon \delta_{i,j}. \quad (5.23)$$

The drift coefficients are related to the deterministic part of the Langevin force and are functions of $\{\theta_j\}$. On the contrary, the diffusion coefficients correspond to the stochastic part of the force and have a constant non zero value only for diagonal elements. Having determined the drift and diffusion terms, the Fokker-Planck equation describing the same stochastic process and equivalent to the Langevin equation (5.12) is expressed as

$$\partial_t W = \sum_{i=1}^M \left[- \frac{\partial}{\partial \theta_i} \left(\frac{\kappa}{\gamma} f_i^{\text{det}} W \right) + \frac{\epsilon}{\Delta z} \frac{\partial^2}{\partial \theta_i^2} W \right]. \quad (5.24)$$

The Fokker-Planck equation is usually not amenable to an analytical solution in a

general case. Nonstationary solutions may be found for specific cases, such as problems involving linear drift and constant diffusion coefficients [Risken 1984]. For the particular case of Eq.(5.24), we restrict our discussion to the stationary regime, for which the solution can be expressed by

$$W_{st} = \mathcal{N} \exp \left\{ -\frac{\kappa}{\gamma} \frac{\Delta z}{2\epsilon} \sum_i \left[\frac{(\theta_{i+1} - \theta_i)^2}{(\Delta z)^2} - \frac{2\beta}{\sqrt{1 - r \sin^2 \theta_i}} \right] \right\}, \quad (5.25)$$

as can be easily seen by substitution. $\mathcal{N}$ is a normalization constant which guarantees that the probability density integrated over all possible states is unity. The form of the right-hand side of this expression indicates that the steady-state probability for the system to be in a state $\{\theta_j\}$ is related to the free energy of the liquid crystal in this state. To write expression (5.25) in terms of the free energy, we first define a discretized form of F , deduced from the continuous form given by (5.3).

$$F_M = \frac{A\kappa}{2} \sum_i \left\{ \frac{(\theta_{i+1} - \theta_i)^2}{(\Delta z)^2} - \frac{2\beta}{\sqrt{1 - r \sin^2 \theta_i}} \right\} \Delta z, \quad (5.26)$$

where the index M stands for the number of points considered in the spatial discretization. This form of the free energy leads to the correct deterministic model when the Euler-Lagrange equations are derived in the continuum limit. With this expression of F , the steady-state solution (5.25) of the Fokker-Planck equation reads

$$W_{st} = \mathcal{N} \exp \left[-\frac{1}{A\gamma\epsilon} F_M \right]. \quad (5.27)$$

ϵ characterizes the correlations of the thermal noise. Noticing that the canonical stationary distribution of the configuration $\{\theta_j\}$ is given by $W_{st} = \mathcal{N} \exp(-F/kT)$, where k is the Boltzmann constant and T is the temperature of the liquid crystal, the value of ϵ can be obtained as $\epsilon = kT/A\gamma$.

Expression (5.27) shows that in steady state, the free energy plays the role of the generalized potential $U(\theta)$ introduced in Section 1. Information on the statistical distribution of the different modes in a stationary regime can then be directly deduced by studying the variations of F as the input intensity is increased above threshold. Absolute probabilities cannot be determined, but ratios between probabilities to observe a given asymptotic state can be worked out by comparing the free energy of the liquid crystal in these states. To illustrate this discussion, we return to the simplified model of a small dielectric anisotropy described in Chapter III, which allows us to write an analytical expression of the molecular orientation θ .

In the limit of small r , the different deterministic solutions for θ have been discussed previously (Section III.1), and were found to be described in steady state by the successive modes (3.6) of the elliptic sine function. With this form of the solution θ , an analytical expression of the free energy can be deduced,

$$\mathcal{F} = A\kappa\beta \int_0^d dz \left\{ \frac{r}{2} \frac{1}{\mu^2 \left[1 - \frac{1}{\mu^2} \operatorname{sn}^2 \left(\sqrt{(\beta r)z} \right) \right]} \operatorname{cn}^2 \left(\sqrt{(\beta r)z} \right) \operatorname{dn}^2 \left(\sqrt{(\beta r)z} \right) - \left[1 - \frac{r}{\mu^2} \operatorname{sn}^2 \left(\sqrt{(\beta r)z} \right) \right]^{-1/2} \right\}, \quad (5.28)$$

where $1/\mu$ is the modulus of the elliptic functions introduced in Chapter III. The functions $\operatorname{cn}(u, 1/\mu)$ and $\operatorname{dn}(u, 1/\mu)$ can be related to the Jacobian elliptic sine function through the relations $\operatorname{cn}^2(u, 1/\mu) = 1 - \operatorname{sn}^2(u, 1/\mu)$ and $\operatorname{dn}^2(u, 1/\mu) = 1 - 1/\mu \operatorname{sn}^2(u, 1/\mu)$ [Byrd and Friedman 1954].

For intensities below threshold, the free energy is simply expressed by the linear relation $F = -\kappa\beta d$. Performing the integral numerically for β above threshold, the free energy is found to be a decreasing function of the input intensity. It has a negative sign, since the electromagnetic energy overcomes the elastic energy for intensities above threshold. For a fixed value of the mode index, the free energy is a decreasing negative function of the intensity, confirming the fact that the potential minimum is deeper when β is increased. Furthermore, for a fixed intensity, the free energy is more negative the lower the index of the mode, implying that the fundamental mode of the molecular reorientation is the most likely to appear

asymptotically.

The free energy plays the role of the generalized potential $U(\theta)$ introduced in Section 1. The possible modes of reorientation found in the preceding Section can be discussed now in terms of their respective free energies. Each possible mode of the solution corresponds to a minimum of $F(\theta)$; the minimum is deeper for the fundamental one, and its value increases with the order of the mode. When a transition occurs to a higher mode, the state of the system corresponds to a local minimum of the potential, the absolute minimum corresponding always to the fundamental mode of the nonhomogenous reorientation. The higher modes are consequently only transient solutions, and will evolve asymptotically towards the fundamental reorientation under the effect of the noise.

An interesting comparison can be performed between the asymptotic probabilities for a transition to different possible modes. We introduce a simplified notation, describing by $W(n)$ the steady-state probability to find the n^{th} mode of the molecular reorientation; the ratio of probabilities to observe a transition to two different modes n and m of the molecular reorientation is proportional to

$$\frac{W(n)}{W(m)} \simeq \exp[-F(n) + F(m)] , \quad (5.29)$$

where $F(n)$ expresses in the same way the free energy of the liquid crystal in the n^{th} mode of the molecular reorientation. The criterion for the n^{th} mode to be more probable than the m^{th} one, is that the corresponding $F(n)$ is lower than $F(m)$. The logarithm of the ratios $W(n)/W(m)$ is plotted in Fig. 5.6 for (a) the first two modes and (b) the second and third modes. These quantities are always positive, and increase for increasing values of the input field, confirming that the lowest order mode is always the more probable to appear when a Freedericksz transition occurs.

In conclusion, a stochastic description of the Freedericksz transition involving internal fluctuations of the molecular orientation shows clearly that higher mode solutions can be reached. Higher modes appear in a numerical simulation for high enough intensities of the laser, even if the fundamental mode is the most likely to appear asymptotically. The characteristic time of a transition is function of the noise

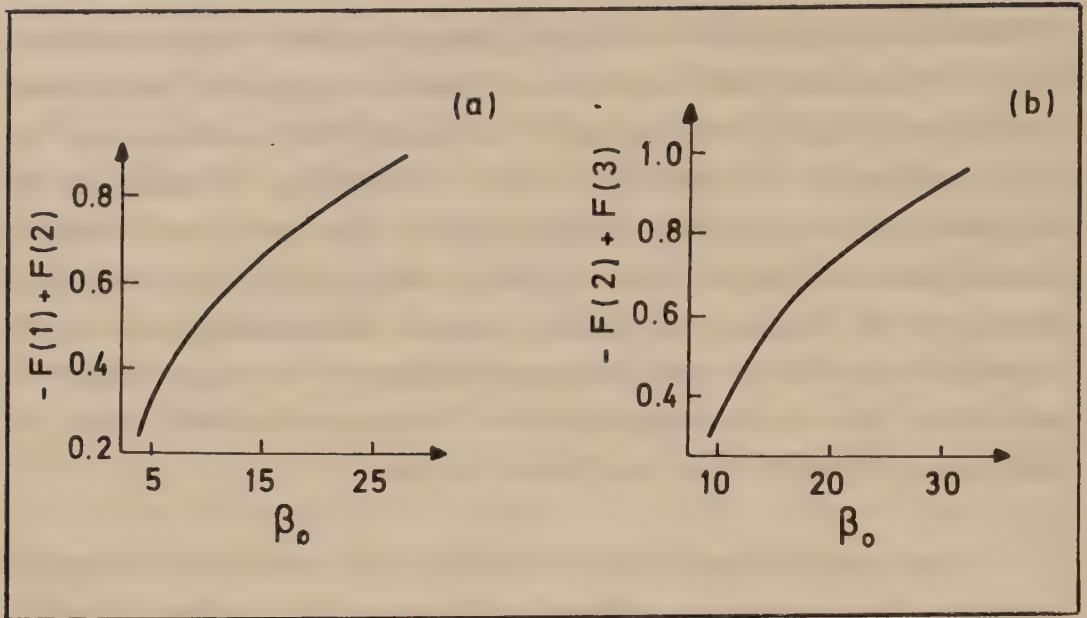


Fig. 5.6 Logarithm of the probability ratios $W(n)/W(m)$ for the first three modes of the nonhomogenous molecular reorientation as a function of the laser intensity. (a) $\ln(W(1)/W(2))$. (b) $\ln(W(2)/W(3))$.

correlations, and is an increasing function of the diffusion constant. It will be interesting in future work to introduce external fluctuations of the laser intensity [Sagues and San Miguel 1985], and to analyze the effects of the resulting colored noise on the optical Freedericksz transition. As already mentioned, colored noise modifies the form of the the potential U driving the dynamics of the transition. One would expect that the characteristic times should then be reduced and the statistics modified.

VI. OPTICAL BISTABILITY IN A FABRY-PEROT INTERFEROMETER CONTAINING A NEMATIC LIQUID CRYSTAL

VI.1 OPTICAL BISTABILITY

Optical Bistability is a nonlinear effect resulting in hysteresis of the light transmitted by an optical system, two different states of the transmission being possible for a single value of the light intensity driving the system. Any system presenting a nonlinear optical property can under appropriate conditions become bistable when a feedback mechanism is provided. In many practical cases, the feedback mechanism is provided by an optical resonator. We review in this Section the general behaviour of a nonlinear optical resonator filled with a passive nonlinear medium.

We consider first the case of an empty Fabry-Perot cavity illuminated by an incident field E_0 . R and T are the intensity reflection and transmission coefficients of the two end mirrors ($R+T = 1$). For an empty resonator, assuming that the fields can be approximated by plane waves, the internal field E is related to the incident field in steady state by

$$E = \sqrt{T} \frac{E_0}{1 - R \exp(i\Phi_l)} , \quad (6.1)$$

Φ_l is the linear phase shift corresponding to the detuning between the incident light frequency and the closest eigenmode of the cavity. The transmitted field is given by $E_t = \sqrt{T} E$. When the detuning is zero, the transmitted field is equal to E_0 , the light is transmitted through the resonator. In this situation, a considerable amount of light is stored inside the resonator, the internal field being equal to $\sqrt{T} E_0$. This stored light provides a feedback mechanism that can lead to bistability when a nonlinear medium is placed inside the cavity.

To study this effect, we now introduce a dispersive nonlinear medium inside the cavity. The light propagating through the cavity induces a variation of the refractive index of the nonlinear medium. This in turn induces a nonlinear phase shift Φ_{nl} of the light beam. The internal field is then given by

$$E = \sqrt{T} \frac{E_0}{1 - R \exp(i\Phi_\ell + i\Phi_{nl})} . \quad (6.2)$$

To obtain this equation which is strictly valid only in steady state, we have assumed that the round-trip time of light in the cavity is much shorter than the response time of the medium. We assume for simplicity the nonlinear medium to be a Kerr medium, whose refractive index is simply given by $n = n_0 + n_2 I$, where I is the internal intensity. In this case Φ_{nl} is a linear function of the intensity, $\Phi_{nl} = \chi I$, where χ is a proportionality constant. Multiplying the fields by their complex conjugates, we obtain the relation between the internal and input intensities

$$I = \frac{T I_0}{1 + R^2 - 2R \cos(\Phi_\ell + \chi I)} . \quad (6.3)$$

In the case of a high finesse resonator, only one cavity mode is important and we can expand the cosine for small arguments about the resonance for this mode. We obtain

$$I = \frac{1}{T} \frac{I_0}{1 + \frac{R}{T^2} (\Phi_\ell + \chi I)^2} . \quad (6.4)$$

A useful characteristic of the resonator is its transmission $\mathcal{T}$, defined as the ratio of the transmitted intensity to the input intensity

$$\mathcal{T} = \frac{I_t}{I_0} = T \frac{I}{I_0} . \quad (6.5)$$

From Eq.(6.4), $\mathcal{T}$ can be expressed as

$$\mathcal{T} = \frac{1}{1 + \frac{R}{T^2} (\Phi_I + \chi I)^2} . \quad (6.6)$$

This transcendental equation can be solved graphically as a function of the internal intensity I . In Fig. 6.1(a), the resonance curve is $\mathcal{T}(I)$ as given by Eq.(6.6). The straight lines represent the linear relation (6.5) for different values of the input intensity I_0 , corresponding simply to different values of the slope. For a given input intensity, the intersections between the two curves give the possible values of the transmission of the Fabry-Perot. We see that there is a domain of incident intensities for which the transmission becomes multivalued. When the input intensity is low, a single value of the transmission characterizes the resonator. When I_0 is increased, the transmission slowly increases until a threshold value $I_0 = I_d$ where two more intersection points become possible. This is followed by a second threshold at $I_0 = I_u$ where the system has no choice but to jump discontinuously to a point at the right of the resonance peak of $\mathcal{T}$. Increasing further the input intensity leads to a decrease of the transmission.

If the input intensity is now decreased back to a low value, the transmission $\mathcal{T}$ increases until it reaches its maximum value, where a discontinuous jump brings it down again to a low value.

This hysteresis effect is illustrated in Fig. 6.1(b), where the internal intensity is plotted as a function of the input intensity. Arrow 1 labels the discontinuous jump from the lower branch of the solution to the upper branch, occurring when the system jumps from one side of the transmission peak to the other, for $I_0 = I_u$. When the input intensity is decreased from this point, the feedback provided by the resonator is enough to sustain a high intensity inside the cavity, allowing the system to stay on the upper branch. When the upper branch ceases to be a solution, a switch-down occurs, labelled by arrow 2. At this point, the internal intensity is not large enough to maintain the system at resonance.

The stability of the different solutions can be investigated by a linear stability analysis. A general criterion can be formulated only for the part of the curve $I(I_0)$ characterized by a negative slope, which is always unstable. No general criterion can be expressed at this point concerning the parts of Fig. 6.1(b) with positive slope. Furthermore, we have assumed that the internal field follows adiabatically the

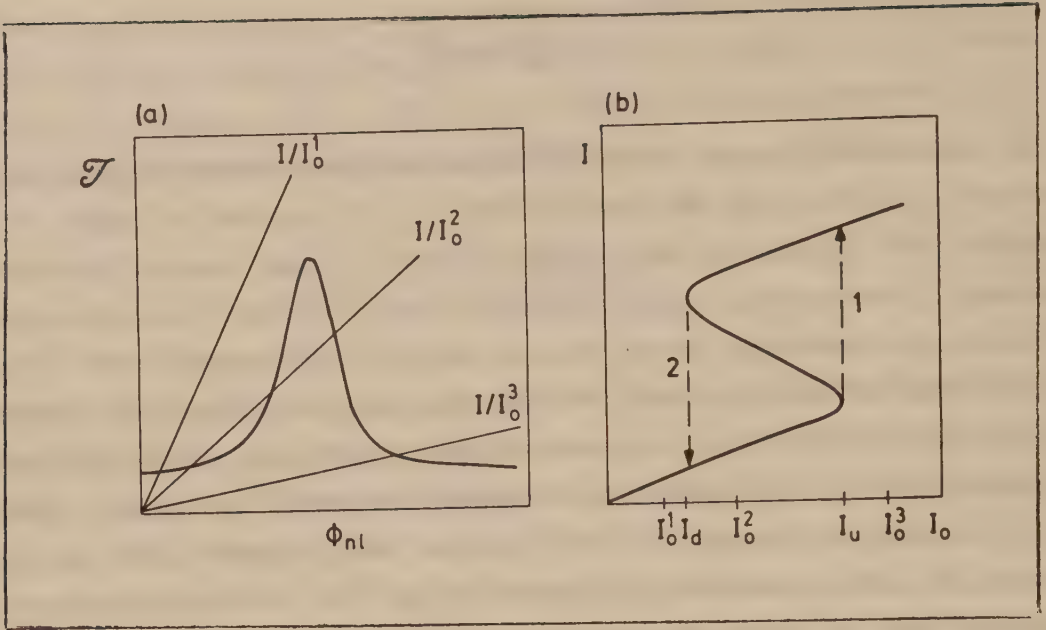


Fig. 6.1 (a) Transmission curve of a Fabry-Perot resonator containing a nonlinear medium. The straight lines display I/I_o . (b) Internal intensity as a function of the input intensity. $I_{o,1} < I_d < I_{o,2} < I_u < I_{o,3}$.

variations induced in the medium, and that a Debye-type relaxation characterizes the dynamics of the nonlinearity, completely described by a single dynamical equation. Under these assumptions, instabilities of the type discussed by Ikeda [Ikeda 1979, Ikeda et al. 1980] can not be induced in this system, no chaotic regime can be found.

We study in the next Section the behaviour of a Fabry-Perot cavity for the specific case where the nonlinear medium is a nematic liquid crystal sample, and the source of the nonlinearity is the light induced molecular reorientation. In this case, reorientation is induced in the medium for internal intensities larger than the threshold of the OFT. Below threshold, the cavity is characterized by a linear regime. Above threshold, the nonlinear induced phase shift leads to a nonlinear relationship between the input and the output intensities. Bistable behaviour of the cavity can then be found under certain conditions.

VI.2 BISTABILITY NEAR THE OPTICAL FREEDERICKSZ TRANSITION

Due to light induced molecular reorientation, a NLC placed inside a Fabry-Perot cavity is expected under some conditions to induce a bistable output of the resonator. The problem of a Fabry-Perot cavity filled with a NLC is however different from usual dispersive optical bistability in a nonlinear resonator, because of the combined effects of the first and second-order phase transitions present in this case. The molecular reorientation occurring when the internal intensity is larger than the threshold is a second-order phase transition, coupled in the present system to a first-order phase transition imposed by the feedback mechanism in the resonator. New effects resulting from this less conventional system are discussed in this Section.

We consider a homeotropically aligned nematic liquid crystal of thickness d placed inside a Fabry-Perot cavity, and irradiated by a CW laser of intensity β , linearly polarized along the direction $\mathbf{e}_x$, and propagating along the easy direction $\mathbf{e}_z$ of the medium perpendicularly to the resonator mirrors and cell walls. For an internal intensity larger than the threshold of the OFT, a nonlinear phase shift Φ_{nl} is experienced by the light due to light induced molecular reorientation in the medium. It is expressed for the geometry at hand by

$$\Phi_{nl} = \frac{2\pi}{\lambda} \int_0^d dz (n_{eff} - n_0) , \quad (6.7)$$

as discussed in Chapter IV.

We approximate the incident field by a plane wave and neglect the transverse inhomogeneities of the molecular reorientation. Furthermore, we limit our discussion to values β of the internal intensity that remain close to the OFT threshold β_{th} , to avoid further complications due to the appearance of higher order modes of the director angle field as the incident intensity is increased. The molecular reorientation was discussed for this case in Chapter III; the director angle was approximated by $\theta(z) = a \sin(\pi z/d)$, and 3 solutions were found for a , namely $a = 0$ and $a = \pm a_\infty$, with a_∞ given by (3.30). With this form of the solution, the

nonlinear phase shift becomes

$$\Phi_{nl} = s \frac{\beta - \beta_{th}}{\beta}, \quad (6.8)$$

where

$$s = \frac{2\pi n_0 r d}{3\alpha\lambda} \Theta(\beta - \beta_{th}). \quad (6.9)$$

Θ is the Heaviside function, $\Theta(x) = 0$ for $x < 0$ and 1 for $x > 0$. The Θ function reflects the fact that no reorientation is induced if the internal intensity is lower than β_{th} . The dependence of Φ_{nl} on the internal intensity is illustrated in Fig. 6.2.

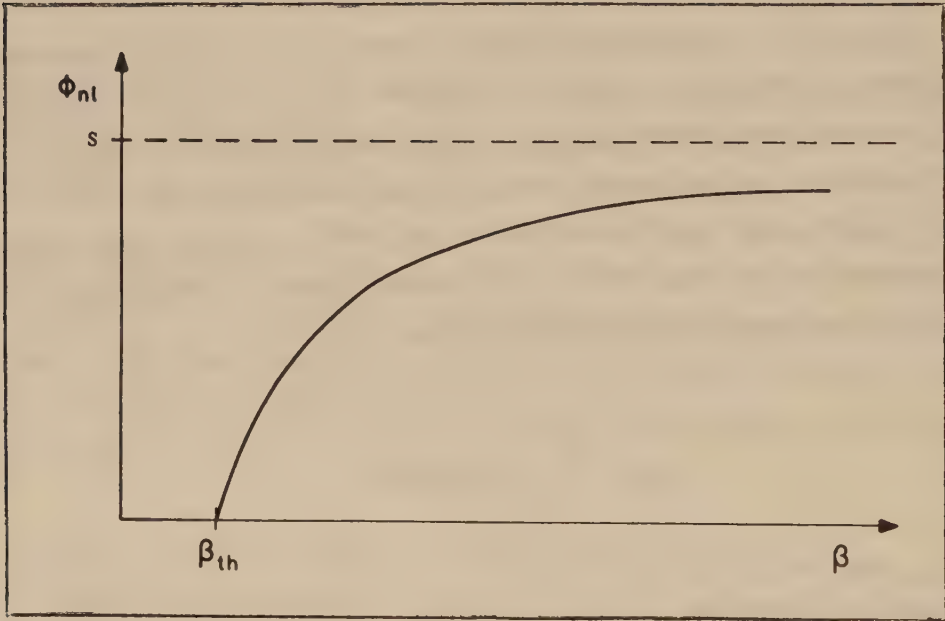


Fig. 6.2 Nonlinear phase shift in function of the input intensity.

When the NLC is placed inside a Fabry-Perot resonator, the internal intensity is governed by Eq.(6.4), which can be rewritten using the expression (6.7) of the nonlinear phase shift as

$$\beta = \frac{1}{T} \frac{\beta_0}{1 + \frac{R}{T^2} \left[\Phi_l + s \left[1 - \frac{\beta_{th}}{\beta} \right] \right]^2} . \quad (6.10)$$

Below threshold, $s = 0$ and Eq.(6.10) yields a linear relationship between internal and incident intensities

$$\beta = \frac{1}{T} \frac{\beta_0}{1 + \frac{R}{T^2} \Phi_l^2} . \quad (6.11)$$

Above threshold, the linear solution is always possible, since $a = 0$ is a possible solution for the molecular reorientation. The linear solution is represented by the curve (L) in Fig. 6.3. Two further angle distributions become possible above threshold, but they lead to the same nonlinear phase shift, since they correspond to the physically undistinguishable angles θ and $-\theta$. In contrast to more conventional situations in optical bistability, this nonlinear solution yields a quadratic relation between β and β_0

$$\beta = \frac{1}{T} \frac{\beta_0 + \frac{2sR}{T} \beta_{th}(\Phi_l + s) \pm \sqrt{\Delta}}{2 \left[1 + \frac{R}{T^2} (\Phi_l + s)^2 \right]} , \quad (6.12)$$

where

$$\Delta = \beta_0^2 + \frac{4sR}{T} \beta_{th} [\beta_0(\Phi_l + s) - sT\beta_{th}] . \quad (6.13)$$

The nonlinear solution (6.12) is illustrated in Fig. 6.3 (curve (N)). The condition $\Delta > 0$ imposes a condition on the existence of the nonlinear branch of the solution, expressed by

$$\beta_0 > 2s\beta_{th} \left[\sqrt{\frac{R^2}{T^2} (\Phi_l + s)^2 + R} - \frac{R}{T} (\Phi_l + s) \right] = \beta_{0,C} \quad (6.14)$$

This mathematical condition should not be confused with the threshold of the OFT, which occurs at

$$\beta_{0,\text{th}} = T\beta_{\text{th}} \left[1 + \frac{R}{T^2} \Phi_l^2 \right]. \quad (6.15)$$

For the solution (6.12) to be physical, β must be larger than the threshold of the OFT. Two situations are then possible, represented in Fig. 6.3. (a) displays the case where the intersection between L and N corresponding to the threshold point $(\beta_{0,\text{th}}, \beta_{\text{th}})$ belongs to the nonlinear upper branch, implying that the nonlinear solution only has a physical existence for values of the input intensity larger than $\beta_{0,\text{th}}$. In this case the internal intensity is always a single valued function of the incident intensity, linear for $\beta_0 < \beta_{0,\text{th}}$ and nonlinear for larger values of β_0 . Case (b) displays the situation where multiple solutions are possible. The intersection between L and N corresponding to the threshold point occurs in the lower nonlinear branch. In this case, the domain limited by $\beta_{0,\text{c}} \leq \beta_0 \leq \beta_{0,\text{th}}$ is characterized by 3 possible solutions. We emphasize that the situation is different from usual bistability, since only two of these solutions are related to the nonlinear process induced by the light in the optical medium. Another difference with usual bistability is the presence in all cases of a domain characterized by 2 solutions, for $\beta_0 \geq \beta_{0,\text{th}}$. This is related to the fact that the source of the nonlinearity is characterized by a threshold, implying that for $\beta < \beta_{\text{th}}$ the nonlinear solution has no physical sense, since no molecular reorientation is induced.

We see in Fig. 6.3 that bistability is possible if the linear solution (L) crosses the nonlinear one (N) in its lower branch. This is the case if the critical point $\beta_{0,\text{c}}$ corresponds to a value $\beta \geq \beta_{\text{th}}$. A straightforward calculation leads to the corresponding value of the internal intensity

$$\beta(\beta_{0,\text{c}}) = s\beta_{\text{th}} \sqrt{\frac{R}{T^2 + R(\Phi_l + s)^2}}. \quad (6.16)$$

The condition for bistability is satisfied if one has

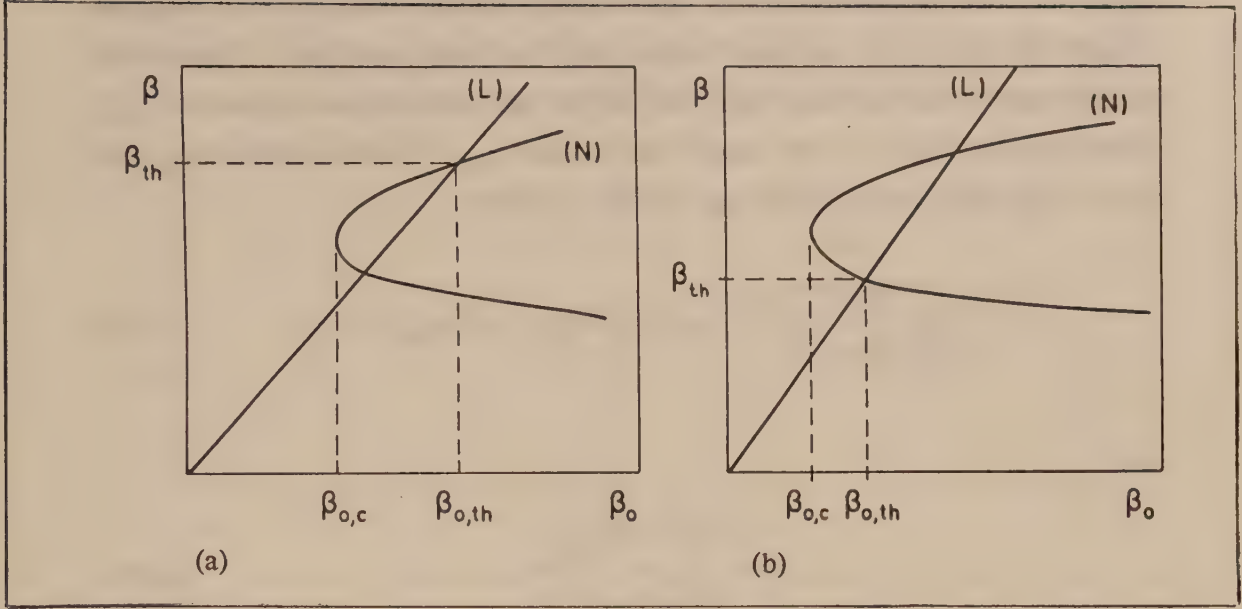


Fig. 6.3 Linear (L) and nonlinear (N) solutions of the resonator equation (6.10), for arbitrary units. The threshold corresponds to the upper (a) or lower (b) intersection of the two curves.

$$s \sqrt{\frac{R}{T^2 + R(\Phi_l + s)^2}} > 1 . \quad (6.17)$$

Expression (6.17) yields directly the possible range of values of the linear detuning for which bistability is possible

$$-s - \sqrt{s^2 - T^2/R} \leq \Phi_l \leq -s + \sqrt{s^2 - T^2/R} . \quad (6.18)$$

Stability analysis

In the regions where multiple solutions are possible, the stability of each solution is studied to determine which is (or are) the stable output(s) of the Fabry-

Perot. The linear stability analysis is easy, but lengthy. Since the same type of calculation was performed several times earlier in this thesis (cf. for example Chapter III, Section 1), we chose to give only the final result here. The stability analysis leads to an eigenvalue equation, and the sign of the largest eigenvalue $\Gamma_{\max}$ determines the stability of the steady-state solution a_{st} , which is stable if $\Gamma_{\max}$ is negative. Its sign is given by the sign of the expression

$$h = \frac{\beta_0}{T + \frac{R}{T^2} (\Phi_l + \Phi_{nl}^0)^2} (1 - \psi a_{st}^2 - 9\alpha a_{st}^2/4 + 3\alpha\psi a_{st}^4/4) - \beta_{th} , \quad (6.19)$$

with

$$\psi = \frac{\frac{2sR}{T} (\Phi_l + \Phi_{nl}^0)}{T + \frac{R}{T} (\Phi_l + \Phi_{nl}^0)^2} . \quad (6.20)$$

and where Φ_{nl}^0 is the nonlinear phase shift corresponding to the non perturbed steady-state configuration.

For the homogenous solution $a_{st} = 0$, we clearly have $\psi = \Phi_{nl} = 0$, which leads to $h = \beta - \beta_{th}$. The zero solution is then stable for $\beta_0 \leq \beta_{0,th}$, and is unstable for higher input intensities.

To study the stability of the non-zero solution, a_{st} has first to be expressed as a function of β_0 , using (3.30) and (6.9) before the sign of h can be checked. For the case where no bistability is possible, the non-zero solution is always stable. When the input intensity is continuously increased from zero, the internal intensity increases first linearly with β_0 , until it reaches the threshold value of the OFT where the nonlinear solution becomes possible. Past this point, the OFT occurs and the zero solution is unstable, a further increase of β_0 leads to a nonlinear increase of β . In the case of bistability, the upper branch characterized by a positive slope is always stable; the part of the lower branch that satisfies $\beta \geq \beta_{th}$ corresponds to an unstable solution. These results are summarized in Fig. 6.4, where only the physical solutions are reported. Unstable regions are represented by dashed lines. Case (a) displays the situation where the criterion (6.18) is not satisfied, implying that no bistability is possible ($\Phi_l = 2$). Case (b) on the other hand satisfies the bistability criterion, $\Phi_l = -10$, multiple solutions are then possible. Arrow 1 corresponds to the

switch-up point, occurring for $\beta_0 = \beta_{th}$, when the linear solution becomes unstable. The system jumps then discontinuously to the upper nonlinear branch. When the input intensity is decreased from this point, the system stays on this branch until β_0 reaches the critical value $\beta_{0,c}$ for which the switch-down to the linear solution occurs, labelled by arrow 2.

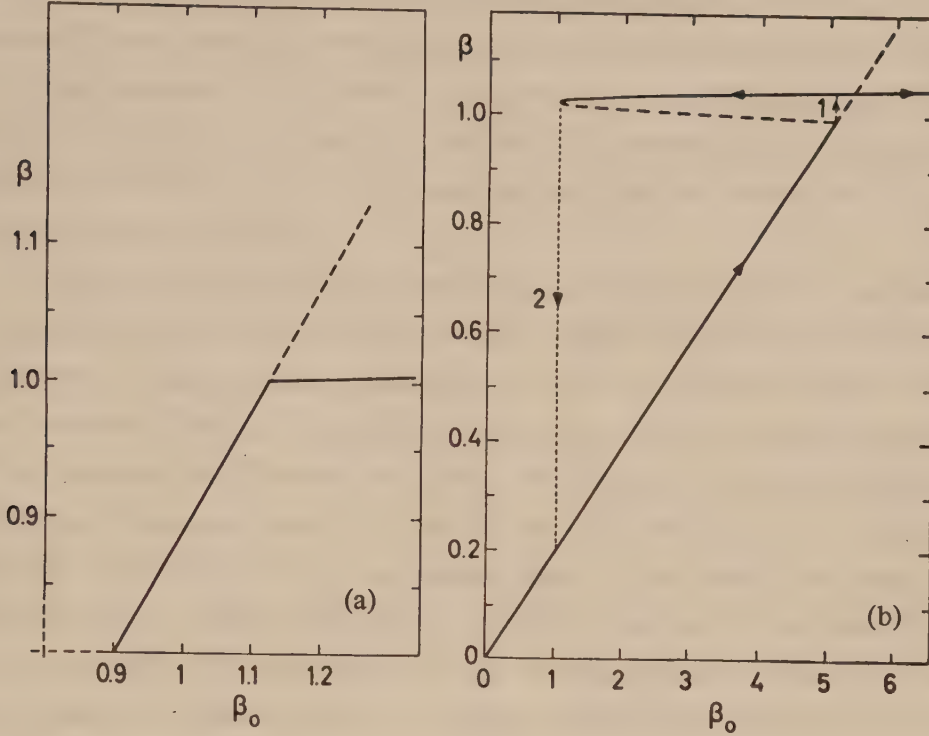


Fig. 6.4 Internal intensity as a function of the input intensity for a Fabry-Perot resonator containing a NLC. (a) $\Phi_l = 2$. (b) $\Phi_l = -10$.

It is important to realize that even if the resonator shows bistable regime due to the nonlinearity of the liquid crystal, the Freedericksz transition does not exhibit any hysteresis. The value of the internal intensity corresponding to the switch down is still higher than the threshold value of the OFT. The case where the Freedericksz transition itself shows hysteresis is not considered here since it can not be described by the model developed in this work, but it could be an interesting problem to be studied in future developments. To study such a system, the one elastic constant approximation must be removed, to allow a first-order Freedericksz transition in the

liquid crystal [Ong 1983]. When the NLC is placed inside the Fabry-Perot cavity, the switches between the different branches of the transmission correspond to first-order phase transitions. In this case, the study of optical bistability leads to an analysis of the coupling between two first-order phase transitions.

VII. SUMMARY AND OUTLOOK

In this thesis, we have studied the molecular reorientation induced by a laser beam in a nematic liquid crystal. Molecular reorientation occurs when the laser intensity is larger than the threshold of the optical Freedericksz transition. It induces a nonlinear variation of the index of refraction of the liquid crystal and affects the propagation of the light through the medium. This process is an interesting nonlinear optical effect that can lead to useful applications in optical devices, like for example all optical logical elements.

In general, the equation describing the light-induced molecular reorientation is not amenable to an analytical solution. A first analytical approximation was discussed for the case of small dielectric anisotropy in a one-dimensional model. It lead to a mode structure for the longitudinal distribution of the molecular reorientation. Several solutions were found to be possible: for intensities just above threshold, the Freedericksz transition occurs to a unique spatial distribution of the reorientation angles. It is described by the fundamental mode of the elliptic sine function. For higher intensities, higher modes become available, described by the higher modes of the elliptic sine. A dynamical study of the molecular reorientation shows that only the fundamental mode is stable. The second analytical approximation discussed the small reorientation regime, in a two-dimensional model where transverse correlations due to elastic molecular forces are taken into account. Inhomogeneous transverse profiles of the reorientation could be found in steady state, even under plane wave pumping.

A numerical solution of the full two-dimensional nonlinear problem for the case of a gaussian transverse profile of the laser intensity confirms the mode structure of the molecular reorientation on the longitudinal dimension, and shows that a similar mode structure characterizes the solution in the transverse dimension. The intensity needed to reach a higher mode increases with the order of the mode. A stability analysis showed again that only the fundamental mode of the molecular reorientation is stable. But for large enough intensities, a higher mode of the molecular reorientation can be sustained in a transient regime for very large times. Furthermore, critical slowing down is exhibited when the intensity is close to threshold, characterizing the second-order phase transition occuring with the

Freedericksz transition. The nonlinear phase shift induced by the variation of the index of refraction in the medium can be measured to distinguish between the different modes of the molecular reorientation.

A stochastic approach to the molecular reorientation was developed in a one-dimensional model, introducing thermal fluctuations in the nonlinear medium. It lead to transitions to higher modes of the molecular reorientation and to tunnelling effects between different modes. This result is important since it shows that higher modes of the reorientation can effectively be reached in a real system involving internal noise. This one-dimensional model can be extended to the two-dimensional case, to include transverse effects in the molecular reorientation. Future work in this direction can yield information on the stability of the higher transverse modes when internal noise is present in the system.

The case of a Fabry-Perot cavity containing a NLC was discussed as an application of our model. Only the lowest mode of the reorientation was considered. Bistable output of the nonlinear resonator was found to be possible in the high finesse approximation. Due to the form of the nonlinearity, the nonlinear branches of the transmission curves of the resonator are solutions of a quadratic equation instead of a cubic one, as it is usually the case in more conventional optically bistable systems. Considering the threshold nature of the Freedericksz transition, a fixed value of the input intensity can correspond to 1, 2 or 3 output regimes.

This problem can be extended in a future work to consider for example the case of a ring resonator containing a NLC illuminated by two counterpropagating beams. The nonlinear medium provides in this case a coupling between the two beams, and various instabilities can possibly occur. In particular, mode coupling can influence the bistable characteristics of the resonator, inducing effects such as crossed-coupled bistability or slaved bistability [Marquis et al. 1986]. This last phenomenon occurs when one mode operates very far from the resonance of its transmission peak, but is forced to jump to a different transmission regime when the second mode is driven through its own transmission resonance peak. Such effects have interesting applications in photonic logic, since they allow to generate a number of logical functions by addressing a nonlinear system by two independent beams.

Another possible extension of our model is to go past the one elastic constant approximation. This would allow first-order phase transitions in the nonlinear medium. Coupling this transition with the first-order phase transition occurring in the resonator when the output switches between possible transmission regimes will be an

interesting direction for future investigations.

A model similar to the one developed in this work to treat the nonlinear optical effects due to molecular reorientation in liquid crystals can be used to discuss the magneto optical properties of thin films of amorphous magnetic media (rare earth/transition metal, such as GdTbCo or TbFeCo) [Shieh and Kryder 1986, Suits et al. 1986]. These media have no atomic orientation, but are characterized by an orientation of their magnetic dipoles. They have a uniaxial magnetic anisotropy and their dipoles can be reoriented in a magnetic or optical field. The magnetization is determined following the same general procedure of minimizing the free energy of the medium. Interesting applications of such materials are developed in the domain of optical data storage, read/write optical disks, etc..., and could be studied with a model similar to the one developed in this work.

Appendix A : Derivation of the electromagnetic energy

The electromagnetic energy density is expressed by

$$F_{em} = 1/2 [E \cdot D + B \cdot H]$$

Using Maxwell's equations with fields written in the form $E \exp(-i(\mathbf{k} \cdot \mathbf{r} - \omega t))$, $E \cdot D$ and $B \cdot H$ become in a plane wave model

$$E \cdot D = E \cdot \left[\frac{1}{i\omega} \nabla \times H \right] = E \cdot \left[\frac{1}{i\omega} (-i\mathbf{k} \times H) \right] = -\frac{1}{c} E \cdot [\mathbf{n}_p \times H]$$

$$B \cdot H = \mu_0 H \cdot \left[\frac{1}{\mu_0 i\omega} \nabla \times E \right] = \frac{1}{c} H \cdot [\mathbf{n}_p \times E] = -\frac{1}{c} E \cdot [\mathbf{n}_p \times H]$$

The last equality is verified for a non conducting medium, without current. Adding these two contributions, one obtains

$$F_{em} = -\frac{1}{c} E \cdot [\mathbf{n}_p \times H] = \frac{1}{c} [E \times H] \cdot \mathbf{n}_p$$

Expressing this energy in terms of the Poynting vector defined by $\mathbf{S} = E \times H$, yields the final expression (2.6)

$$F_{em} = \frac{1}{c} \mathbf{S} \cdot \mathbf{n}_p$$

Appendix B : Nonhomogenous solution of Eq.(3.28)

A first integration of Eq.(3.28) yields

$$(d_x a)^2 = C - \sigma a^2 + \xi a^4/2$$

where the integration constant C is given by the asymptotic condition

$$d_x a(x = \pm\infty) = d_x a_\infty = 0,$$

which leads to

$$C = \sigma a_\infty^2 + \xi a_\infty^4/2$$

with a_∞ expressed by (3.30). Writing σ in terms of a_∞ and ξ leads to

$$d_x a = \sqrt{\xi/2} (a_\infty^2 - a^2)$$

Choosing the positive sign in the expression of a_∞ , the two solutions of this equation are written as

$$a(x) = a_\infty \tanh\left[a_\infty \sqrt{\xi/2} x + C_1\right]$$

$$a(x) = a_\infty \coth\left[a_\infty \sqrt{\xi/2} x + C_2\right]$$

where C_1 and C_2 are arbitrary constants of integration.

REFERENCES.

- A.M.Albano, N.B.Abraham and D.E.Chyba, *Am. J. Phys.* **52**, 161, 1984.
- S.M.Arakelian, A.S.Karayan and Yu.S.Chilingaryan, *Opt. Spectr. USSR* **55**, 298, 1983.
- S.I.Ben Abraham, *Phys. Rev.* **A14**, 1251, 1976.
- P.F.Byrd and M.D.Friedman, "Handbook of Elliptic Integrals for Engineers and Physicists", Springer Verlag, Berlin, 1954.
- M.Born and E.Wolf, "Principles of Optics", Pergamon Press, Oxford, 1970.
- M.M.Cheung, S.D.Durbin and Y.R.Shen, *Opt. Lett.* **8**, 39, 1983.
- L.Csillag, I.Janossy, V.F.Kitaeva, N.Kroo and N.N.Sobolev, *Mol. Cryst. Liq. Cryst.* **84**, 125, 1982.
- S.D.Durbin, S.M.Arakelian and Y.R.Shen, *Phys. Rev. Lett.* **47**, 1411, 1981a.
- S.D.Durbin, S.M.Arakelian and Y.R.Shen, *Opt. Lett.* **6**, 411, 1981b.
- S.D.Durbin, S.M.Arakelian and Y.R.Shen, *Opt. Lett.* **7**, 145, 1982.
- P.C.Fife, "Mathematical Aspects of Reacting and Diffusing Systems", Springer Verlag, Berlin, 1970.
- F.C.Frank, *Discuss. Faraday Soc.* **25**, 19, 1958.
- V.Freedericksz and A.Repiewa, *Z.Phys.* **42**, 532, 1927.
- P.G.de Gennes, "The Physics of Liquid Crystals", Clarendon Press, Oxford, 1975.
- L.A.Goodman, in "Introduction to Liquid Crystals", edited by Priestley, Wojtowicz and Sheng, Plenum Press, New-York, 1974.
- H.Hsiung, L.P.Shi and Y.R.Shen, *Phys. Rev.* **A30**, 1453, 1984.
- K.Ikeda, *Opt. Comm.* **30**, 257, 1979.
- K.Ikeda, H.Diado and O.Akimoto, *Phys. Rev. Lett.* **45**, 709, 1980.
- F.J.Kahn, *Appl. Phys. Lett.* **20**, 199, 1972.
- P.P.Karat and N.V.Madhusudana, *Mol. Cryst. Liq. Cryst.* **36**, 51, 1976.
- A.J.Karn, S.M.Arakelian, Y.R.Shen and H.L.Ong, *Phys. Rev. Lett.* **57**, 448, 1986.
- I.C.Khoo, *Phys. Rev.* **A25**, 1636, 1982.
- I.C.Khoo, *Phys. Rev.* **A27**, 2747, 1983.
- I.C.Khoo and J.Y.Hou, *JOSA* **B2**, 761, 1985.
- L.Landau and E.Lifshitz, "Quantum Mechanics", Pergamon Press, Oxford, 1977.
- O.Lehmann, *Wied. Ann.* **40**, 401, 1890.

- O.Lehmann, Annln. Phys. 2, 649, 1900.
- F.Marquis, P.Dobiasch, P.Meystre and E.M.Wright, JOSA B3, 50, 1986.
- H.L.Ong, Phys. Rev. A28, 2393, 1983.
- H.L.Ong, Phys. Rev. A31, 3450, 1985, and Appl. Phys. Lett. 46, 822, 1985.
- C.W.Oseen, Trans. Faraday Soc. 29, 883, 1933.
- H.Risken, "The Fokker-Planck Equation : Methods of Solutions and Applications", Springer Series in Synergetics, Springer Verlag, Berlin, 1984.
- F.Sagues and M.San Miguel, Phys. Rev. A32, 1843, 1985.
- Y.R.Shen, Phil. Trans. R. Soc. Lond. A313, 327, 1984.
- H.P.D.Shieh and M.H.Kryder, Appl. Phys. Lett. 49, 473, 1986.
- J.C.Suits, R.H.Geiss, C.J.Lin, D.Rugar and A.E.Bell, Appl. Phys. Lett. 49, 419, 1986.
- O.Svelto, "Principles of Lasers", Plenum Press, New-York, 1976.
- B.Ya.Zel'dovich, N.V.Tabiryan and Yu.S.Chilingaryan, Sov. Phys. JETP 54, 32, 1981.
- B.Ya.Zel'dovich and N.V. Tabiryan, Sov. Phys. JETP 55, 656, 1982.
- H.Zocher, Trans. Faraday Soc. 29, 945, 1933.

Acknowledgements

I gratefully acknowledge the encouragement and aid of Prof. P.Meystre, whose support (and sometimes teasing) led to the accomplishment of this work.

I am grateful to Prof. H.Walther, who gave me the possibility in the course of my graduate studies at the Max-Planck Institut für Quantenoptik to perform the last phase of my work at the Optical Sciences Center of the University of Arizona, and to Prof. R.R.Shannon for his invitation to stay at the OSC.

I am indebted to Prof. Y.R.Shen for suggesting the subject of my dissertation and getting me interested in the physics of Liquid Crystals; and to Drs. E.M.Wright, J.Gea-Banacloche, P.Dobiasch and Prof. A.E.Kaplan for very helpful discussions at different stages of this work.

I am grateful to all the people who helped to create a friendly atmosphere, both at the MPQ and at the OSC, especially to the group of the "Theoretical Container" of the MPQ, and to Ken for helping me with the English.

My thanks to Frau Pfister, for her efficient drawings.

I acknowledge partial financial support from the Swiss National Fund for Scientific Research.

Finally, none of this work would have been accomplished without the constant encouragement of Claude, whom I want to thank here for his invaluable help and patience during all the good and the bad moments of my work.

Personal data

Name: Fabienne Dominique Marquis

Date of birth: 21. November 1961

Place of birth: Sion (Switzerland)

Citizenship: Swiss

Marital status: single

Address: Max-Planck Institut für Quantenoptik
D-8046 Garching bei München

Resume

1967 - 1972: Elementary School, Sion

1972 - 1979: College classique S.M.A., Sion
"Maturité" in June 1979

1979 - 1984: Undergraduate studies at the Ecole Polytechnique Fédérale, Physics Department, Lausanne (Switzerland). "Diplôme" at the Institut de Physique Théorique on "Phononic effects in optical transitions", under the supervision of Professor A.Quattropani.

1984 - 1986: Graduate studies at the Ludwig-Maximilians-Universität München; Dissertation at the Max-Planck Institut für Quantenoptik under

the supervision of Professor P.Meystre.

April - October 1986:

Research assistant at the Optical Sciences
Center of the University of Arizona, Tucson
(USA).

